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REDUCIBLE AND IRREDUCIBLE COMPONENTS OF THE pH VALUE IN DILUTE AQUEOUS SOLUTIONS OF SODIUM CHLORIDE

Main attention is paid to the definition of the reducible and irreducible components of pH in aqueous salt solutions and to the determination of their temperature and concentration dependences. It is shown that the temperature dependence of the reducible pH component in dilute aqueous solutions of sodium chloride has a linear character and differs from that in pure water only by the value of its slope, which increases, as the salt concentration grows. At the same time, the temperature dependence of the irreducible pH component is non-monotonic and has a minimum at the temperature that is optimal for the human and mammalian life activities (36.6 °C). The existence of a characteristic salt concentration dividing the family of the temperature dependences of pH into two subfamilies with different behaviors of their temperature dependences has been established.

Keywords: aqueous solution, sodium chloride, pH, reducible component, irreducible component.

1. Introduction

The role of pH in the human or mammalian life has been known for long [1, 2]. However, the question “Why does this occur?” still remains unanswered. In our recent work [3], we posed and answered the question about a close connection between the boundaries of the life activity for humans and many mammals and the corresponding pH values of their blood.

In work [4], it was proved that the relaxation time $\tau(\text{pH}, T)$ of non-equilibrium pH values acquires the minimum values at the temperature and pH values belonging to the vital activity region, i.e., the corresponding rates of biophysical processes substantially exceed their rates in any other (pH, T) region. The

points (pH_0, T_0) where $\tau(\text{pH}, T)$ is minimum coincide with the corresponding optimal temperatures for the human or mammalian vital activity. Based on the above considerations, it was found that $T_o = 36.6$ °C, $T_l = (30 \pm 2)$ °C, and $T_u = 42$ °C, where the subscripts “o”, “l”, and “u” denote the optimal temperature of the pH relaxation time and the extreme of lower and upper points of the vital activity interval, respectively. The pH values at the optimal point were expectedly found to be somewhat lower than the experimental ones, because the influence of macromolecules, such as albumin, on the pH value was not taken into consideration.

In this paper, we continue the study of the pH value. More specifically, we determine the reducible and irreducible pH components and analyze their dependence on the temperature and the salt concentration in aqueous sodium chloride (NaCl) solutions. The main focus is concentrated on the salt concentrations that are close to those in human blood plasma. It will be proved that the temperature dependence of the irreducible pH component is characterized by a

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point in a vicinity of which the pH value has a local minimum, and which coincides with the optimal temperature for the human life activity.

2. Experimental Part

In our studies, we used freshly distilled water of purity class II according to DSTU ISO 3696:2003. It was obtained using an Adrona Crystal EX Double Flow water purification system (Adrona SIA, Latvia). The 0.9 wt% aqueous NaCl solution of pharmaceutical grade for infusions (Darnytsya, Ukraine) was used as the primary one.

Experimental pH measurements were carried out in a temperature interval of (294÷323) K and for the NaCl solution concentrations corresponding to $\zeta = (180\div 1620)$, where ζ is the number of water molecules per salt ion. The indicated ζ -values correspond to conventional solution concentrations $\omega = (0.9\div 0.1)$ wt%. It is easy to see that ζ and the concentration ω are related by the relation $\zeta = (1-\omega)/\omega$.

The solutions were prepared gravimetrically using a Radwag AS 220.R2 balance (Radwag, Poland). The total relative error of mass measurements did not exceed 0.05%. The temperature was controlled by means of an ultra-thermostat UTU-10 (Krakow, Poland) to within an error of ± 0.1 K.

The pH value was measured in accordance with the IUPAC recommendations [5, 6] on an AZ Bench Top Water Quality Meter 86505 (AZ Instrument Corp., Taiwan) equipped with a temperature-sensitive probe. The total relative error of pH measurements was 0.5% [7, 8]. Before the measurements, the pH meter was calibrated using reference buffer solutions with pH 4.00 and 7.00 at a temperature of 298 K.

The same temperature stabilization and measurement conditions were provided for all examined solutions. The peculiarities of the method used when measuring the pH in NaCl solutions contacting with atmospheric carbon dioxide were described in detail in works [3, 4].

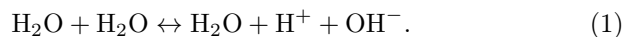
In work [7], it was shown that the absolute error of pH measurements using a pH meter with two-point calibration, automatic temperature compensation, and a combined glass electrode cannot be better than ± 0.02 . The main problem arising when calculating the uncertainty of instrumental pH measurements are the residual liquid junction potential

and the following phenomena: asymmetry potentials, mixing effects, memory effects, temperature effects (e.g., temperature gradients above the electrode), diaphragm clogging, electrical noise (cable shielding), cross-contamination, and carbonate exchange (in alkaline calibration buffers) [7, 8]. In work [9], it was shown that if all the above-mentioned uncertainty factors are taken into account when measuring the pH value in water and dilute aqueous solutions in a pH interval of 3.5–7.5, the absolute pH measurement error reaches a value of $\pm(0.10\div 0.14)$.

In work [10], it was shown that the purity of applied reagents and the knowledge of exact solution concentrations have a significant impact on the determination of the solution pH via instrumental measurements.

3. Reducible and Irreducible Contributions to the pH of Aqueous NaCl Solutions

In general, pH values are governed by both the temperature effects and the presence of salt. The thermal motion of molecules obeys the Maxwell distribution, which guarantees the existence of such binary collisions that can destroy the integrity of at least one molecule and release a cation H^+ ,



Note that the mechanism of binary collisions between water molecules, as was shown in work [4], leads to the following contribution to the pH value:

$$pH = -\lg c_H^+ \quad (2)$$

where

$$c_H^+ = \frac{2}{\sqrt{\pi}} \sqrt{\frac{3T_D}{2T}} \exp\left(-\frac{3T_D}{2T}\right) \quad (3)$$

is the concentration of hydrogen cations, and T_D is the effective dissociation temperature of water molecules in the aqueous medium. It is easy to see that

$$pH(T) = pH(T_{tr}) - \frac{1}{2} \lg \frac{T_{tr}}{T} - \frac{3T_D}{2T_{tr}} \left(1 - \frac{T_{tr}}{T}\right) \lg e, \quad (4)$$

where

$$pH(T_{tr}) = -\lg \frac{2}{\sqrt{\pi}} - \frac{1}{2} \lg \frac{3T_D}{2T_{tr}} + \frac{3T_D}{2T_{tr}} \lg e. \quad (5)$$

As one can see from Fig. 1, the pH values calculated for water at the fixed dissociation temperature $T_D = 2400$ K using formula (4) correlate quite satisfactorily within the measurement error with the experimental data obtained in a temperature interval of $(273 \div 333)$ K.

In order to put the calculated and experimental pH values in agreement for water in the whole temperature interval, where the liquid phase exists, it is necessary to introduce a weak temperature dependence of T_D . Such an introduction finds its natural explanation in the framework of the approach presented in work [4]. When salts are added to water, water molecules dissociate with the formation of hydrate shells around them [12, 13]. As a result,

- substantial electric fields of cations and anions reduce the dissociation threshold $T_D(\zeta)$ in the binary collision model [4];
- a mechanism of additional dissociation due to considerable fluctuations in the intensity of electric fields inside the instantaneous hydrate shell becomes activated.

Accordingly, in the first case, the temperature dependence of the corresponding contribution to the pH is linear in the temperature,

$$\text{pH}_{\text{red}}(T, \zeta) = \text{pH}(T_*(\zeta), \zeta) + A(\zeta) [T - T_*(\zeta)] + \dots, \quad (6)$$

as it occurs for water,

$$\text{pH}_w(T) = \text{pH}_w(T_*) + A_w (T - T_*) + \dots \quad (7)$$

Here, $T_*(\zeta)$ is a calibration point, like $T_w = T_*(\zeta \rightarrow \infty) = 25$ °C for water. The reducible pH component must satisfy the following properties:

$$\lim_{\zeta \rightarrow \infty} \text{pH}_{\text{red}}(T, \zeta) = \text{pH}_w(T), \quad (8)$$

$$\lim_{\zeta \rightarrow \infty} A(\zeta) = A_w, \quad (9)$$

$$\lim_{\zeta \rightarrow \infty} T_*(\zeta) = T_w. \quad (10)$$

The pH contributions of the second type differ from the first-case contributions by their concentration dependence. In particular, at $\zeta \rightarrow \infty$,

$$\lim_{\zeta \rightarrow \infty} \text{pH}_{\text{irr}}(T, \zeta) = 0. \quad (11)$$

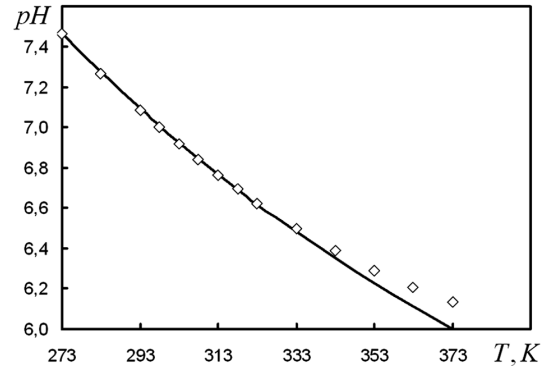


Fig. 1. Temperature dependence of pH for water: diamonds denote experimental values [11] and the solid curve was plotted according to formula (4) with $T_D = 2400$ K

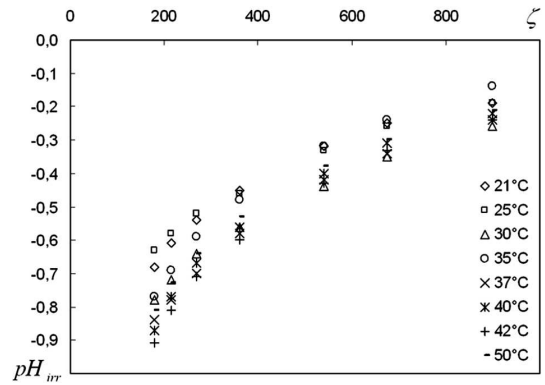


Fig. 2. Concentration dependences of the irreducible pH component $\text{pH}_{\text{irr}}(T, \zeta)$ at various temperatures for aqueous solutions of sodium chloride

From Eq. (11), it follows that, at relatively low salt concentrations,

$$\text{pH}_{\text{irr}}(T, \zeta) \approx \frac{\zeta_1}{\zeta} + \dots \quad (12)$$

The concentration dependence of $\text{pH}_{\text{irr}}(T, \zeta)$ was calculated using the formula

$$\text{pH}_{\text{irr}}(T, \zeta) = \text{pH}(T, \zeta) - \text{pH}(T, \zeta_0), \quad (13)$$

where $\text{pH}(T, \zeta)$ are the experimental pH values obtained in works [3, 4], and ζ is the number of water molecules per NaCl ion. It was assumed that the pH values for the salt solution at the concentration $\zeta_0 = 1620$ and pure water are practically identical. The behavior of $\text{pH}_{\text{irr}}(T, \zeta)$ as a function of the concentration is shown in Fig. 2.

Assuming that, in general, $\text{pH}(T, \zeta)$ is the sum

$$\text{pH}(T, \zeta) = \text{pH}_{\text{red}}(T, \zeta) + \text{pH}_{\text{irr}}(T, \zeta), \quad (14)$$

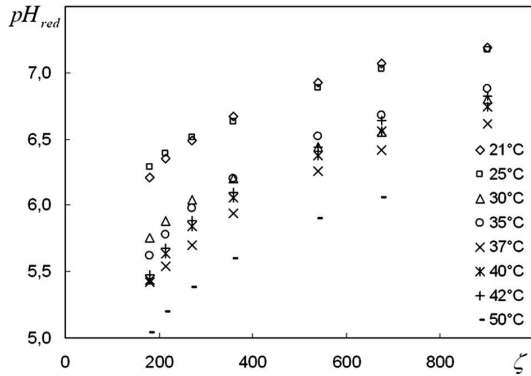


Fig. 3. Concentration dependences of the reducible pH component $\text{pH}_{\text{red}}(T, \zeta)$ at various temperatures for aqueous solutions of sodium chloride

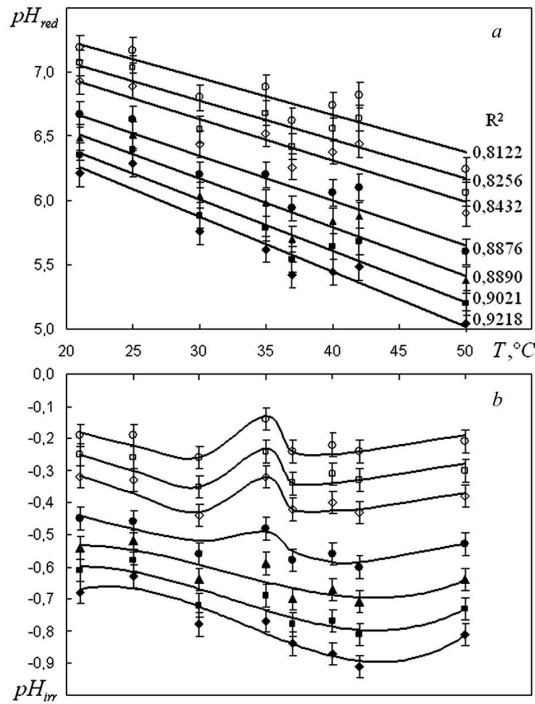


Fig. 4. Temperature dependences of $\text{pH}_{\text{red}}(T, \zeta)$ (a) and $\text{pH}_{\text{irr}}(T, \zeta)$ (b) for aqueous NaCl solutions with various salt concentrations $\zeta = 180$ (◆), 215 (■), 270 (▲), 360 (●), 540 (◇), 675 (□), and 900 (○). The determination coefficients R^2 for the linear approximations of $\text{pH}_{\text{red}}(T, \zeta)$ are shown in panel a

we obtain the following formula for the concentration dependence of $\text{pH}_{\text{red}}(T, \zeta)$ (Fig. 3):

$$\text{pH}_{\text{red}}(T, \zeta) = \text{pH}(T, \zeta) - \text{pH}_{\text{irr}}(T, \zeta). \quad (15)$$

In many respects, the temperature dependences of the $\text{pH}_{\text{red}}(T, \zeta)$ and $\text{pH}_{\text{irr}}(T, \zeta)$ values for aqueous

NaCl solutions at fixed concentrations are more informative (Fig. 4). As one can see from Fig. 1, the behavior of the temperature dependence of $\text{pH}_{\text{red}}(T, \zeta)$ is close to linear. This fact confirms our expectation that $\text{pH}_{\text{red}}(T, \zeta)$ values are governed by binary collisions, but with a lower dissociation temperature. The different character of the temperature dependence of $\text{pH}_{\text{irr}}(T, \zeta)$ testifies to a deeper relation between the irreducible pH component and the peculiarities in the behavior of living organisms.

Let us additionally illustrate the sense of the division of a physical quantity into the reducible and irreducible components using the polarizability of the nitrogen molecule N_2 as an example. The polarizability α_{N_2} of the nitrogen molecule is often approximated by the expression

$$\alpha_{\text{N}_2} \approx \alpha_{\text{N}} + \alpha_{\text{N}}. \quad (16)$$

Approximation (16) can be made accurate, if we add a contribution $\delta\alpha(r_{12})$ that arises owing to the interaction between the nitrogen atoms and depends on the distance r_{12} between them,

$$\alpha_{\text{N}_2} = \alpha_{\text{N}} + \alpha_{\text{N}} + \delta\alpha(r_{12}), \quad (17)$$

where the term $\delta\alpha(r_{12}) \rightarrow 0$ at $r_{12} \rightarrow \infty$. In this connection, it is common to call the quantity $\alpha'_{\text{N}_2} = 2\alpha_{\text{N}}$ the reducible component of the polarizability α_{N_2} , and the additional contribution $\delta\alpha(r_{12})$ the irreducible component of α_{N_2} .

4. Important Features in the Temperature Dependences of $\text{pH}_{\text{red}}(T, \zeta)$ and $\text{pH}_{\text{irr}}(T, \zeta)$ for Aqueous NaCl Solutions

In this section, our attention will be focused on the following issues:

- the temperature dependence of the slope of the lines approximating the concentration behavior of $\text{pH}_{\text{red}}(T, \zeta)$;
- the concentration dependence of calibration points;
- a relation between the temperature dependence of $\text{pH}_{\text{irr}}(T, \zeta)$ and the physiological features of the human and mammalian life activities.

The concentration dependence of the coefficient $A(\zeta) = d(\text{pH}_{\text{red}})/dT$, which describes the slope of the lines $\text{pH}_{\text{red}}(T, \zeta)$ and which was determined from the experimental data depicted in Fig. 4, is shown in Fig. 5. Note that it was additionally assumed that the

temperature dependence of the tangent of the slope $A(\zeta)$ changes more substantially than the calibration temperature does. From the analysis of Fig. 4, *a*, it follows that if $\text{pH}_{\text{red}}(T, \zeta)$ is approximated by linear dependences, the value of the determination coefficient R^2 decreases, as ζ increases.

The numerical values of $A(\zeta)$ for NaCl solutions with various values of ζ and the corresponding value for pure water, $A_w = A(\zeta \rightarrow \infty)$, are compared in Table 1 and Fig. 5. As one can see, the limiting formula (9) is satisfied, but the limit value A_w is approached slowly.

From the analysis of Fig. 4, it follows that the limiting relation (11) is also fulfilled satisfactorily. It is easy to verify that the temperature-averaged values of $\text{pH}_{\text{irr}}(T, \zeta)$ from Fig. 4 satisfy the equation

$$\text{pH}_{\text{irr}}(T, \zeta) \approx \frac{\zeta_1}{\zeta} + \frac{\zeta_2}{\zeta^2} + \dots, \quad (18)$$

where $\zeta_1 \approx 178$ and $\zeta_2 \approx 4417$ (Fig. 6), for each analyzed concentration, as it should be in accordance with Eq. (12).

At the same time, another circumstance is more important. Namely, the curves $\text{pH}_{\text{irr}}(T, \zeta)$ can be grouped into two subfamilies with different curve structures: for $180 < \zeta < \zeta_*$ and $\zeta_* < \zeta < 900$, where $\zeta_* \approx 300$. The minimum values of the curves $\text{pH}_{\text{irr}}(T, \zeta)$ from the first subfamily are observed just in a vicinity of the temperature $T_u \approx 42$ °C, which, according to work [3], has the meaning of an upper limit for the temperature interval of the human and mammalian vital activities.

On the other hand, the minimum values of $\text{pH}_{\text{irr}}(T, \zeta)$ for the curves from the second subfam-

Concentration dependence of the slope of the straight lines $\text{pH}_{\text{red}}(T, \zeta)$ for aqueous NaCl solutions

ζ	$A(\zeta)$
∞	-0.013
900	-0.029
675	-0.031
540	-0.032
360	-0.035
270	-0.038
215	-0.040
180	-0.043

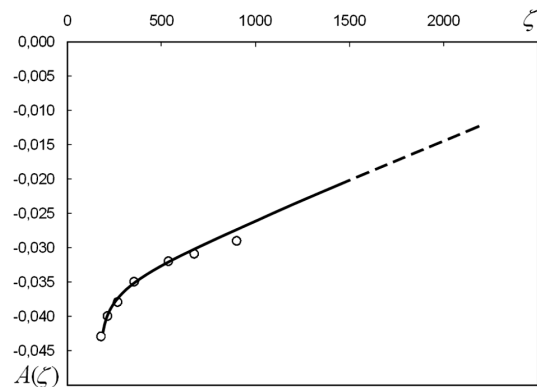


Fig. 5. Concentration dependence $A(\zeta)$ for aqueous NaCl solutions

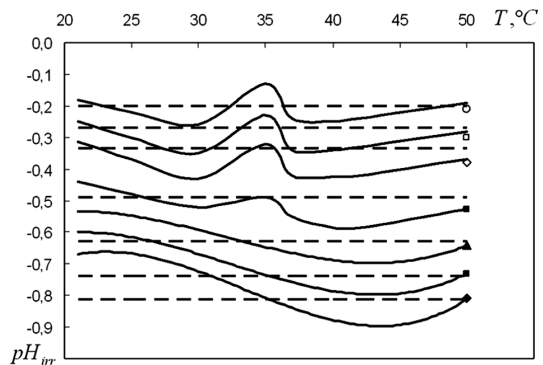


Fig. 6. Temperature dependences of $\text{pH}_{\text{red}}(T, \zeta)$ for aqueous NaCl solutions with various salt concentrations $\zeta = 180(\blacklozenge)$, 215 (\blacksquare), 270 (\blacktriangle), 360 (\bullet), 540 (\diamond), 675 (\square), and 900 (\circ). Solid curves correspond to the corresponding curves in Fig. 4, *b*; dashed linear dependences were plotted according to Eq. (18)

ily are observed at the points $T_l^{\text{min}} \approx 30$ °C and $T_o^{\text{min}} \approx 37$ °C corresponding to the lower limit of the life activity interval and the optimal temperature of the human and mammalian life activities, respectively [14–24].

5. Conclusions

The pH value for dilute aqueous solutions of sodium chloride is a sum of two components: reducible and irreducible. The reducible component depends linearly on the temperature and, therefore, cannot be responsible for the specific details of the human and mammalian life activities. Therefore, the main parameters of the latter have to be characterized by the irreducible pH component, which is an order of magnitude smaller than the reducible component.

The minimum points in the curves of the irreducible pH component for aqueous sodium chloride solutions are directly related to the temperatures that can be interpreted as the lower ($T_l \approx 30^\circ\text{C}$) and upper ($T_u \approx 42^\circ\text{C}$) limits of the temperature interval of the human and mammalian life activities. At the same time, the minimum in a vicinity of the temperature $T_o \approx 37^\circ\text{C}$ can be interpreted as the optimal temperature for the vital activity of humans and mammals.

The proportionality between the irreducible pH component value and the sodium chloride concentration may testify that minor pH changes are associated with corresponding changes in the salt composition of blood plasma that affect the state of human organism.

Since the reducible and irreducible pH components behave themselves differently, of particular importance becomes the issue concerning the conformational modifications of albumin [25] and other proteins, which depend on the pH value. The addition of proteins will change, first of all, the reducible pH component, and the conformational modification of albumin will be primarily induced by the electric fields of hydrogen cations, which, surely, will directly affect the irreducible pH component as well.

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**ЗВІДНА ТА НЕЗВІДНА ЧАСТИНИ
ВОДНЕВОГО ПОКАЗНИКА рН РОЗБАВЛЕНИХ
ВОДНИХ РОЗЧИНІВ ХЛОРИДУ НАТРІЮ**

Основна увага в статті приділяється означенню звідної та незвідної складових водневого показника рН та визна-

ченню їх температурних і концентраційних залежностей у водно-сольових розчинах. Показано, що в розбавлених водних розчинах хлориду натрію температурна залежність звідної складової рН має лінійний характер, що відрізняється від аналогічної залежності у воді тільки нахилом ліній температурної залежності рН, який зростає зі зростанням концентрації солі. Незвідна складова рН є немонотонною за температурою і приймає мінімальне значення за оптимальної температури життєдіяльності людини і ссавців (36,6 °C). Встановлено існування характерної концентрації солі, яка поділяє сімейство температурних залежностей рН на дві підсистеми з різними характерами залежності від температури.

Ключові слова: водний розчин, хлорид натрію, водневий показник, звідна частина, незвідна частина.