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RESEARCH OF THE DEPENDENCES OF DIELECTRIC PROPERTIES AND FREQUENCY SPECTRA OF DIELECTRIC LOSSES IN NaCl AQUEOUS SOLUTION ON THE STATE PARAMETERS

The frequency spectra of dielectric permittivity, $\varepsilon_1(\nu)$, and dielectric loss, $\varepsilon_2(\nu)$, coefficients for the aqueous solution of sodium chloride and their dependences on the concentration, density, and temperature have been studied. Numerical calculations of those parameters are made for a certain choice of the potential interaction energy $\Phi_{ab}(r)$ and the radial distribution function $g_{ab}(r)$. The results obtained are in satisfactory agreement with experimental data.

Keywords: frequency spectrum, dielectric permittivity, dielectric losses, intermolecular interaction energy, radial distribution function, friction coefficient, relaxation time.

It is known that, in order to describe the dielectric properties of condensed media, one must know such field characteristics as the external electric field strength E and the electric induction D, which are related in the case of constant fields by means of the static dielectric permittivity coefficient ε_s. However, if the applied electric field periodically changes in time, $E(t)$, the induction also changes periodically in time, $D(t)$, with a certain phase lag. In this case, the relation between those field characteristics includes the complex dielectric permittivity coefficient $\tilde{\varepsilon}(\omega)$ depending on the frequency ω. Its real part is the dielectric permittivity coefficient $\varepsilon_1(\omega)$, and the imaginary one the coefficient of dielectric losses $\varepsilon_2(\omega)$. Plenty of works were devoted to the experimental and theoretical researches of those coefficients for electrolyte solutions.

In works [1, 2], the theory of dielectric permittivity $\varepsilon_1(\omega)$ and dielectric losses $\varepsilon_2(\omega)$ for homogeneous dielectric media under the action of both static and time-dependent external electric fields not depending on spatial coordinates was expounded in detail. The Debye formulas for the frequency dependence $\tilde{\varepsilon}(\omega)$ were obtained in various models, as well as the analytic expressions for $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$. The Debye theory is based on the hypothesis that the equilibrium state is established in time following the exponential law.

In works [3, 4], the transfer phenomena in and the dielectric properties of electrolyte solutions were studied both experimentally and theoretically. The phenomenological theory prevailed in studying the physics of insulators, although the role of microscopic theory grows permanently. In the physics of insulators, the methods of molecular-kinetic theory, temporal autocorrelation functions, and the method of collective variables are widely applied [5–9]. In the cited works, the static and dynamic coefficients $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ were determined.

Experimental data on the dielectric permittivity of aqueous electrolyte solutions were generalized in work [10]. The permittivity as a function of the concentration at radio and ultrahigh frequencies was studied. Relations for the correlation factor of molecules in liquids containing chain associates were obtained in work [11]. Those formulas describe the interrelation between the dielectric properties of liquid phases and their molecular structure. The found expressions can also be used to determine thermodynamic characteristics of the association reaction in liquid phases. The detailed analysis of experimental data concerning the dielectric parameters of the concentrated sodium chloride solution [12] showed that they are described in the framework of the Cole–Cole relaxation model

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rather than the Debye one, which was used earlier. The Debye type of a polar liquid is known to be a special case of the more general Cole–Cole relaxation model.

In work [13], the static dielectric permittivity and the dielectric spectra of 1,3-propanediol and its binary solutions with H-propanol in a frequency range from 9.4 to 73.4 GHz were studied, as well as the density in a wide temperature interval. The frequency dispersion of the dielectric permittivity $\varepsilon(\omega)$ in diole and its solutions was revealed in the examined frequency, ω, and temperature, T, intervals. A discrete spectrum of dipole relaxation times was found. The molecular nature of the concentration dependences revealed for the dielectric properties of the solutions was discussed.

The data on the variation of the dielectric permittivity coefficients $\varepsilon_1(\omega)$, $\varepsilon_2(\omega)$, $\varepsilon_{\infty 1}$, and $\varepsilon_{\infty 2}$ for water and the times of dielectric relaxation $\tau_{\rm D}$ in a temperature interval of 60–200 °C can be found in work [14]. In the cited work, the nonlinearity in the dependence of $\ln \tau_{\rm D}$ on the inverse temperature was revealed. The results of calculations using the Debye–Stokes formula were found to agree well with experimental data within the whole temperature interval.

In the last decades, a lot of works were devoted to studying the concentration, C, and temperature, T, dependences of the frequency dispersion of the dielectric permittivity coefficient $\varepsilon_1(\omega)$ and the spectrum of dielectric losses $\varepsilon_2(\omega)$ in aqueous electrolyte solutions; in particular, these are works [15–20]. In works [7, 15], it was shown that, by analyzing the frequency spectrum of the dielectric permittivity, it is possible to detect the formation of ionic pairs in a solution. With the help of the dielectric spectroscopy method, the formation of ionic pairs in the solutions of cobalt, beryllium, copper, nickel, aluminum, scandium, and other sulfates was revealed at a concentration of 0.05 mol/l. The second Debye region can be easily distinguished in the Cole–Cole diagrams and in the frequency dependences of the dielectric permittivity and dielectric loss coefficients [9, 16, 17]; it manifests itself in the frequency interval of 0.1–2 GHz. On the basis of those data, one can determine the molecular parameters of ionic pairs. The corresponding analysis makes it possible to calculate the relaxation times and the dipole moments of ionic pairs.

While studying the structural and molecular-kinetic state of water in solutions, the method of UHF dielectric spectroscopy was selected in work [18]. The researches were carried out in the frequency interval corresponding to the dispersion maximum for the dielectric permittivity of water. Accordingly, a measurement complex for the experimental determination of $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ in a frequency range of 7–25 GHz was created. A regular research of the $\varepsilon_1(\omega)$- and $\varepsilon_2(\omega)$-dependences in this frequency interval was performed for 60 water–electrolyte systems, including multicomponent and saturated solutions, in wide temperature and concentration intervals. The parameters describing the process of dipole relaxation of water molecules in electrolyte solutions (the static dielectric constant ε_s, the dielectric relaxation time τ, and the enthalpy and entropy of relaxation process activation) were determined.

In work [19], with the help of an UHF dielectrograph and with the use of the point-by-point measurement method, the behavior of the dielectric permittivity in aqueous electrolyte solutions at high concentrations was studied. An anomalous frequency dependence was found at a frequency of 9.1 GHz; namely, the dielectric permittivity of the solution grows and exceeds the dielectric permittivity of water. The concentration and temperature dependences and the dielectrograms of the anomalous behavior of the dielectric permittivity $\varepsilon_1(\omega)$ for a number of aqueous solutions of halogenides of alkali metals, hydrohalic acids, and alkalis were obtained.

In works [20, 21], an anomalous behavior of the solution glycerol–water was researched, as well as the specific characteristics of the kinematic shear viscosity of water. The density of the glycerol aqueous solution was measured along the coexistence curve with the help of the pycnometer method in the temperature interval 293 K $\leq T \leq$ 363 K and the glycerol concentration interval 0.225 mol/l $\leq C \leq$ $\leq$ 0.552 mol/l. The growth of the glycerol–water solution density was demonstrated to depend on the concentration. At the concentration $C = 0.33$ mol/l, this curve has a maximum. This concentration was found to correlate with the maximum of the isobaric expansion coefficient. The dominating role in the determination of the kinematic shear viscosity of water, ν, belongs to the law of corresponding states and the Hilbert principle. It is found that the kinematic viscosity ν is a sum of the argon-like viscosity contribution and the contribution ν_H given by hydrogen bonds. The application of the Hilbert method allowed the parameter ν_H to be determined for the basic characteristic of the hydrogen bond network, i.e. the number of hydrogen bonds per molecule.

In work [22], it was shown that, for a cell filled with a NaCl aqueous solution, the dependence of its resistance on the alternating current frequency cannot be explained as a result of polarization phenomena taking place only at the electrode–electrolyte interface. A physical mechanism was proposed, which explains the monotonic increase of the solution conductivity at frequencies below 10^4 Hz and its constant value in the frequency interval of 10^4–10^5 Hz. The temperature dependences of the diffusion coefficients for Na^+ and Cl^- ions in the NaCl aqueous solutions were calculated, as well as the size of the physically infinitesimal volume in this electrolyte, in which the local equilibrium is established. The space-time behavior of parameters in the NaCl aqueous solution was analyzed. A relationship between the ratios of the alternating current period to certain time characteristics, on the one hand, and the frequency dependence of the specific conductivity in this electrolyte, on the other hand, was established.

Main applications of the dielectric relaxation spectroscopy method to the study of the structure and dynamics of electrolyte solutions are discussed in work [23]. The cited authors also analyzed the nature of ionic solvation and association in solutions. The results obtained are in the good agreement with experimental data obtained by other methods, in particular, the numerical simulation.

In work [24], on the basis of kinetic equations for the one- and two-particle distribution functions of ions, an analytical expression was derived for the complex-valued conductivity $\tilde{\sigma}(\omega)$ of electrolyte solutions. In works [25, 26], the relationship between the complex coefficients of dielectric permittivity $\tilde{\varepsilon}(\omega)$ and specific conductance $\tilde{\sigma}(\omega)$ were used to obtain analytic expressions for the dynamic coefficients of dielectric permittivity $\varepsilon_1(\omega)$ and dielectric losses $\varepsilon_2(\omega)$. In work [26], numerical calculations of the friction coefficients β_a and β_b, the relaxation times τ_a and τ_b for ions of kinds a and b in the momentum space and their relaxation time τ_{ab} in the configuration space, and the dielectric permittivity coefficient $\varepsilon_1(\omega)$ were carried out for three concentrations C of the sodium chloride aqueous solution, in wide intervals of density ρ, temperature T, and frequency ω, and for a certain choice of the particle-to-particle interaction potential $\Phi_{ab}(r)$ and the radial distribution function $g_{ab}(r)$.

This work is aimed at studying the frequency spectrum of the dielectric loss coefficient $\varepsilon_2(\omega)$ and carrying out its numerical calculations for a sodium chloride aqueous solution with three concentrations C in wide intervals of density ρ, temperature T, and frequency ω. As a start point, we use analytic expressions for the coefficients of dielectric permittivity $\varepsilon_1(\omega)$ and dielectric losses $\varepsilon_2(\omega)$, which were obtained in work [26]:

$$\varepsilon_1(\omega) = \varepsilon_\infty + \sum_a \frac{\sigma_a^0 \tau_a}{\varepsilon_0[1+(\omega\tau_a)^2]} \times \\ \times \left[1 + \sum_b \left(\frac{1-(\omega\tau_a)^2 \frac{\tau_{ab}}{\tau_a}}{1+(\omega\tau_a)^2\left(\frac{\tau_{ab}}{\tau_a}\right)^2} A_{ab}(0) \right)\right], \qquad (1)$$

$$\varepsilon_2(\omega) = \sum_a \frac{(\omega\tau_a)\sigma_a^0 \tau_a}{\varepsilon_0[1+(\omega\tau_a)^2]} \times \\ \times \left[1 + \sum_b \left(\frac{\left(1+\frac{\tau_{ab}}{\tau_a}\right)}{1+(\omega\tau_a)^2\left(\frac{\tau_{ab}}{\tau_a}\right)^2} A_{ab}(0) \right)\right], \qquad (2)$$

where

$$A_{ab}(0) = 6\pi \overset{*}{n}_b q_{ab} \int \frac{\partial^2 \overset{*}{\Phi}_{ab}(r)}{\partial r^2} g_{ab}(r) r^2 dr, \qquad (3)$$

$\sigma_a^0 = \frac{n_a e_a^2}{\beta_a}$; $\tau_a = \frac{m_a}{2\beta_a}$; $\tau_b = \frac{m_b}{2\beta_b}$; $\tau_{ab} = \frac{d_{ab}^2}{kT} \frac{\beta_a\beta_b}{\beta_a+\beta_b}$; $d_{ab} = \frac{1}{2}(d_a+d_b)$; $\overset{*}{n}_b = \frac{\pi}{6} n_b d_{ab}^3$; $q_{ab} = \frac{4}{\pi} \frac{e_b\beta_a - e_a\beta_b}{e_a(\beta_a+\beta_b)}$; $\overset{*}{\Phi}_{ab}(r) = \frac{\Phi_{ab}(r)}{kT}$; $\Phi_{ab}(r)$ is the potential energy of interaction between the structural units of the electrolyte solution; $g_{ab}(r)$ the radial distribution function; m_i, e_i, n_i, d_i, and β_i are the mass, charge, concentration, diameter, and friction coefficient, respectively, for ions of sort i $(i = a, b)$; τ_a, τ_b, and τ_{ab} are the relaxation times in the momentum and configuration spaces; $\mathbf{r}_{ab} = \mathbf{q}_b - \mathbf{q}_a$ is the relative distance; $\mathbf{r} = \mathbf{r}_{ab}/d_{ab}$ is the normalized distance between ions of sort a and b; ε_0 the electric constant; k the Boltzmann constant; T the absolute temperature; $\omega = 2\pi\nu$ is the cyclic frequency, and ν the frequency of the process; and ε_∞ is the value of dielectric permittivity coefficient at $\omega \to \infty$. Formulas (1)–(3) make it possible to study the frequency spectra of the coefficients of dielectric permittivity $\varepsilon_1(\omega)$ and dielectric losses

Table 1. **Dependence of ε_1 on ρ, C, and ν for a NaCl aqueous solution at $T = 298$ K**

C, mol/l	ρ, kg/m^3	ν, GHz								
		[30]	0.0238	0.238	2.38	23.8	190	238	1190	2380
0.40	1011	68.4	63.70	63.70	63.69	62.99	50.02	31.80	7.10	5.55
0.50	1018	66.9	61.58	61.58	61.57	60.87	48.11	30.66	7.01	5.52
0.60	1021	65.4	60.96	60.96	60.95	60.25	47.56	30.37	6.98	5.51
0.73	1025	64.6	60.39	60.39	60.38	59.68	47.08	30.15	6.96	5.51
1.00	1039	60.5	58.60	58.60	58.59	57.89	45.73	29.65	6.90	5.49
4.64	1168		42.55	42.55	42.57	43.90	38.59	25.72	6.56	5.40

Table 2. **Dependence of ε_2 on ρ, C, and ν for a NaCl aqueous solution at $T = 298$ K**

C, mol/l	ρ, kg/m^3	ν, GHz							
		0.0238	0.238	2.38	23.8	190	238	1190	2380
0.35	1011	0.006	0.06	0.65	6.38	24.59	28.88	10.56	5.45
0.50	1018	0.006	0.06	0.63	6.22	23.75	27.64	10.15	5.23
0.60	1021	0.006	0.06	0.63	6.17	23.47	27.27	10.04	5.18
0.73	1025	0.006	0.06	0.62	6.13	23.20	26.93	9.95	5.13
1.00	1039	0.006	0.06	0.60	5.94	22.22	25.98	9.70	4.99
4.64	1168	0.001	0.01	0.06	1.79	16.12	20.74	7.89	4.06

$\varepsilon_2(\omega)$, as well as their dependences on the thermodynamic state parameters. According to Eq. (3), the potential terms $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ are determined in terms of the potential energy of interaction between the structural units of the solution $\Phi_{ab}(r)$ and the radial distribution function $g_{ab}(r)$. For a definite choice of the solution model, those equilibrium structural parameters are known from the literature. We confine the consideration to the spherically symmetric case and select a semiphenomenological osmotic solution model [5, 27] described in the framework of the McMillan–Mayer theory. In this approximation, the solvent is supposed to be a continuous medium with the ionic subsystem moving inside. The contribution of a solvent to the transfer and dielectric parameter coefficients is taken into account by introducing the dielectric permittivity coefficient of the solvent ε_{SS} and the friction coefficients β_a and β_b for ions of sort a and b, respectively.

For carrying out the numerical calculations of $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$, in addition to the known molecular parameters m_i, e_i, n_i, and d_i ($i = a, b$), the friction coefficients β_a and the relaxation times τ_a, τ_b, and τ_{ab} are also required. The latter, in accordance with formulas (1) and (2), are expressed in terms of β_a and β_b. For the latter two, we use the following analytic expressions taken from work [28]:

$$\beta_a^2 = \sum_a \frac{4\pi}{3}\rho_a \sum_b d_{ab} \int\limits_0^\infty \nabla^2\, \Phi_{ab}(r) g_{ab}(r) r^2 dr,$$
$$\beta_b^2 = \sum_b \frac{4\pi}{3}\rho_b \sum_a d_{ab} \int\limits_0^\infty \nabla^2\, \Phi_{ab}(r) g_{ab}(r) r^2 dr, \tag{4}$$

where $\nabla^2 = \frac{1}{r^2}\frac{\partial}{\partial r}(r^2 \frac{\partial}{\partial r})$ is the radial part of the Laplace operator, and $\rho_a = m_a n_a$ and $\rho_b = m_b n_b$ are the density of particles of sorts a and b, respectively.

Following work [28], the potential energy of interaction $\Phi_{ab}(r)$ is adopted as a sum of the Lennard-Jones potential energy and the generalized Debye potential taking the configuration and sizes of ions into account:

$$\Phi_{ab} = \frac{4\varepsilon_{ab}}{\varepsilon_{ss}}(r^{-12} - r^{-6}) + R_{ab}\frac{e^{-\kappa^* r}}{r}, \tag{5}$$

where

$$d_{ab} = (d_{aa} + d_{\ bb})/2, \quad R_{ab} = \frac{f z_a z_b e^2 e^{\kappa^*}}{kT\varepsilon_{ss} d_{ab}(1+\kappa^*)},$$

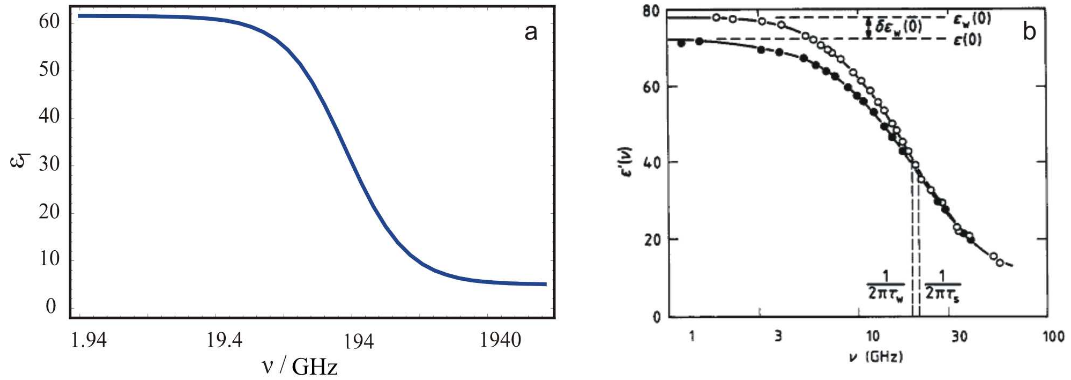

Fig. 1. Frequency spectrum $\varepsilon_1(\nu)$ of a NaCl aqueous solution at $T = 298$ K and $C = 0.5$ mol/l. Formula (1) (*a*); experimental results of work [31]: water (white circles) and sodium chloride aqueous solution (black circles) (*b*)

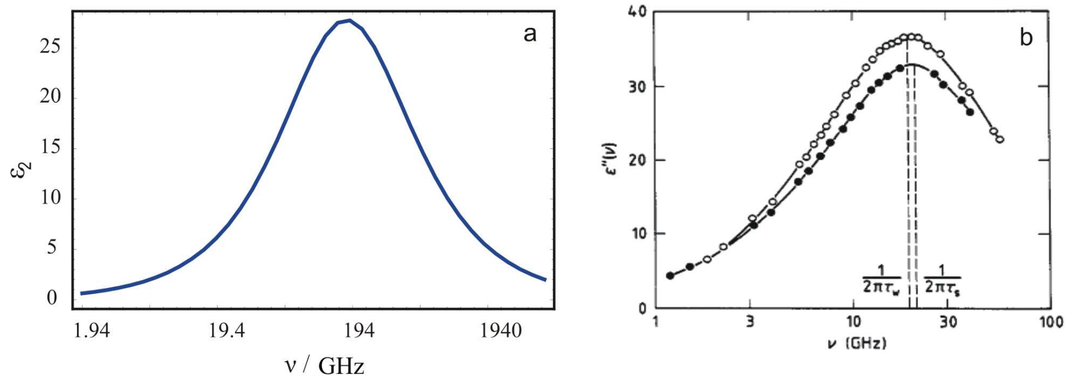

Fig. 2. Frequency spectrum $\varepsilon_2(\nu)$ of a NaCl aqueous solution at $T = 298$ K and $C = 0.5$ mol/l. Formula (2) (*a*); experimental results of work [31] (*b*): water (white circles) and sodium chloride aqueous solution (black circles)

$\varepsilon_{ab} = (\varepsilon_{aa}\varepsilon_{bb})^{\frac{1}{2}}$ is the potential well depth for the particle-to-particle interaction energy, $f = (4\pi\varepsilon_0)^{-1} = 9 \times 10^9$ F/m, ε_0 is the electric constant, ε_{ss} the dielectric permittivity of the solvent, e the elementary charge, d_{aa} and d_{bb} the diameters of ions a and b, respectively, z_a and z_b their valences, and $\kappa^* = \kappa_a d_{ab}$ is the reciprocal Debye screening radius, in which κ is defined in the form $\kappa^2 = \sum_a n_a e_a^2/(\varepsilon_r\varepsilon_0 kT)$ [29]. Following work [5], the function $g_{ab}(r)$ for the ionic subsystem is adopted in the form

$$g_{ab}(r) = y(r, \rho^*)e^{-\frac{\Phi_{ab}(r)}{kT}}, \tag{6}$$

where $y(r, \rho^*)$ is the binary distribution function of two cavities. We restrict the analysis to its contact value $y(r, \rho^*) \approx y(\rho^*)$ at the distance $r = 1$ ($r_{ab} = d_{ab}$) obtained by Carnahan and Starling in the form

$$y(\rho^*) = (2 - \rho^*)/2(1 - \rho^*)^3, \tag{7}$$

where $\rho^* = \pi n d_{ab}^3/6 = \pi\rho d_{ab}^3 N_0/6M$ is the reduced density, ρ the solution density, N_0 the Avogadro constant, and M the molar mass.

On the basis of formula (1) with regard for Eqs. (4)–(7), the numerical calculations were carried out for the friction coefficients β_a and β_b, the relaxation times τ_a, τ_b, and τ_{ab}, and the dielectric permittivity coefficient $\varepsilon_1(\nu)$ as functions of ρ, C, T, and ω. The results of calculations are quoted in work [26]. On their basis, as well as on the basis of formula (2), and taking into account Eqs. (4)–(7), the frequency spectrum $\varepsilon_2(\nu)$ for a NaCl aqueous solution is calculated. The obtained results of numerical calculations are summarized in Tables 1 and 2 and in Figs. 1 to 3.

Tables 1 and 2 contain the numerical values of coefficients of dielectric permittivity $\varepsilon_1(\nu)$ and dielectric losses $\varepsilon_2(\nu)$ in the case of a sodium chloride aqueous

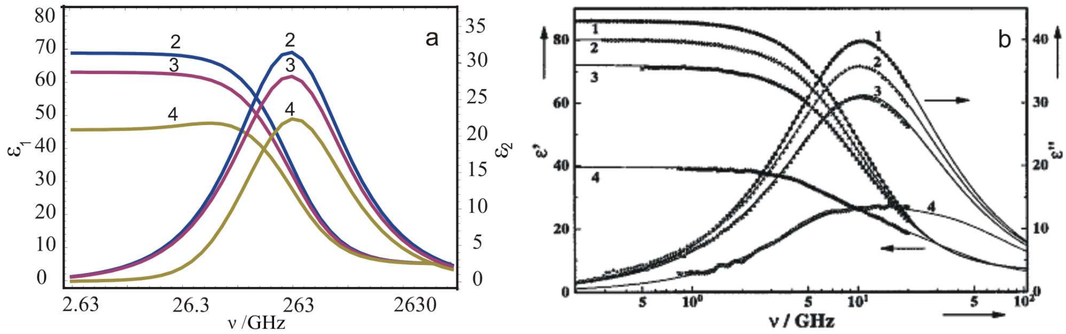

Fig. 3. Frequency spectra $\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ of a NaCl aqueous solution at $T = 298$ K for various concentrations $C = 0$ (*1*, pure water), 0.40 (*2*), 0.99 (*3*), and 4.64 mol/l (*4*). Calculations are made by formulas (1) and (2) (*a*); experimental results of work [32] (*b*)

solution at a temperature of 298 K in the concentration interval 0.35 mol/l $\leq C \leq$ 4.64 mol/l, the density interval 1011 kg/m $\leq \rho \leq$ 1168 kg/m, and the frequency interval 2.38×10^7 Hz $\leq \nu \leq 2.38 \times 10^{12}$ Hz. A comparison of the theoretical results obtained for $\varepsilon_1(\nu)$ with the experimental data of work [30] was made. The behavior of the concentration, density, and frequency dependences of $\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ correspond to experimental results.

In Figs. 1, *a* and 2, *a*, the frequency spectra $\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$, respectively, are depicted in the frequency interval 1.94×10^9 Hz $\leq \nu \leq 1.94 \times 10^{12}$ Hz for the NaCl aqueous solution with a concentration of 0.5 mol/l and at a temperature of 298 K. In Figs. 1, *b* and 2, *b*, the corresponding experimental results [31] for the same values of state parameters and the frequency interval 10^9 Hz $\leq \nu \leq 10^{11}$ Hz are shown. It is evident that the behavior of theoretical and experimental curves for the spectra $\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ coincide.

In Fig. 3, the theoretical values of frequency spectra $\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ in the frequency interval $2.63 \times \times 10^9$ Hz $\leq \nu \leq 2.63 \times 10^{12}$ Hz are exhibited for the sodium chloride aqueous solution at a temperature of 278 K and for three concentrations (0.4, 0.99, and 4.64 mol/l). Figure 3, *b* demonstrates the experimental results of work [32], which were obtained at the same values of state parameters. One can see that the profiles of experimental and theoretical $\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ curves, as well as the corresponding numerical values, are in satisfactory agreement.

Hence, the results of numerical calculations obtained for the selected solution model, intermolecular interaction potential $\Phi_{ab}(r)$, and radial distribution function $g_{ab}(r)$ brought about a satisfactory agreement between theoretical and experimental $\varepsilon_1(\nu)$- and $\varepsilon_2(\nu)$-values for the NaCl aqueous solution in wide intervals of state parameters. The frequency spectra $\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ in a wide frequency range are governed by the dielectric relaxation in the NaCl aqueous solution, which reflects the contribution of relaxation processes.

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ДОСЛІДЖЕННЯ ДІЕЛЕКТРИЧНИХ ВЛАСТИВОСТЕЙ І ЧАСТОТНИХ СПЕКТРІВ ДІЕЛЕКТРИЧНИХ ВТРАТ ВОДНОГО РОЗЧИНУ NaCl ЗАЛЕЖНО ВІД ПАРАМЕТРІВ СТАНУ

Резюме

Досліджено частотні спектри коефіцієнтів діелектричної проникності $\varepsilon_1(\nu)$ і діелектричних втрат $\varepsilon_2(\nu)$ водного розчину хлориду натрію залежно від концентрації, щільності і температури. Зроблено чисельні розрахунки цих коефіцієнтів для водного розчину NaCl при певному виборі потенційної енергії взаємодії $\Phi_{ab}(r)$ і радіальної функції розподілу $G_{ab}(r)$. Отримані результати наведено в таблицях і на малюнках, які знаходяться в задовільній згоді з експериментальними даними.