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КОНСТАНТЫ ЭЛЕКТРОЛИТИЧЕСКОЙ ДИССОЦИАЦИИ СУЛЬФАТОВ ЛИТИЯ, НАТРИЯ И КАЛИЯ В ВОДНО-ЭТАНОЛЬНЫХ РАСТВОРАХ

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Кондуктометрическим методом определены константы электролитической диссоциации сульфатов лития, натрия и калия в водно-этанольных растворах при 25 °С. Li_2SO_4, Na_2SO_4, K_2SO_4 показали снижение их способности к диссоциации. Общепризнано, что степень электролитической диссоциации электролитов в растворе зависит от диэлектрической проницаемости растворителя. Вода и этанол сильно различаются диэлектрической проницаемостью. Поэтому можно получать растворители с различной диэлектрической проницаемостью путем изменения содержания спирта в смеси с водой, таким образом влияя на равновесное состояние соли в растворе. Поэтому в водно-этанольных растворах с увеличением содержания спирта сульфаты лития, натрия и калия должны проявлять свойства слабого электролита. В этом случае зависимость молярной электропроводности от концентрации сульфатов лития, натрия и калия в водно-этанольных растворах будет иметь другой вид. Действительно, с увеличением содержания спирта в водно-этанольных растворах снижается молярная электропроводность вследствие понижения степени электролитической диссоциации. С понижением концентрации соли в растворе возрастает молярная электропроводность, приближаясь к λ_0, как для водного, так и для водно-спиртовых растворов. Как следует из полученных данных, зависимость λ от $C_0(M_2SO_4)$, (M_2SO_4 – начальные концентрации Li_2SO_4, Na_2SO_4, K_2SO_4) для водного раствора с высоким коэффициентом корреляции трансформируется в прямую линию в координатах уравнения Кольрауша, так как сульфаты лития, натрия и калия в водной среде выступают сильными электролитами. Для водно-этанольных растворов высокий коэффициент корреляции в координатах уравнения $4\lambda^3 \cdot C_0(M_2SO_4)^2 = K_{дис} \cdot \lambda_0^3 - K_{дис} \cdot \lambda_0^2 \cdot \lambda$ наблюдается в случае $[C_2H_5OH] > 50$ % объемных. Это означает, что в этих растворах сульфаты лития, натрия и калия проявляют свойства слабого электролита. При содержании спирта от 10% до 40% об. сульфаты лития, натрия и калия выступают электролитом средней силы, и поэтому характерны низкие коэффициенты корреляции для трансформаций в координатах уравнений Кольрауша и $4\lambda^3 \cdot C_0(M_2SO_4)^2 = K_{дис} \cdot \lambda_0^3 - K_{дис} \cdot \lambda_0^2 \cdot \lambda$.

Ключевые слова: водно-этанольные растворы сульфатов лития, натрия и калия, кондуктометрия, константы электролитической диссоциации

CONSTANT OF ELECTROLYTIC DISSOCIATION OF LITHIUM, SODIUM AND POTASSIUM SULPHATES IN AQUEOUS ETHANOL SOLUTIONS

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Constants of electrolytic dissociation of the lithium, sodium and potassium sulphates in aqueous ethanol solutions at 25 °C were determined by conductometric method. Li_2SO_4, Na_2SO_4, K_2SO_4 were shown to decrease their ability to dissociation. It is generally accepted that the degree of electrolytic dissociation of electrolytes in a solution depends on the dielectric constant of the solvent. Water and ethanol differ greatly in their dielectric constant. For this reason, it is possible to prepare the solvent with different dielectric permittivity by changing the alcohol content in a mixture with water, thus, influencing the equilibrium state of the salt in solution. Therefore, in water-ethanol solutions with an increase in the alcohol content, sulfates of lithium, sodium and potassium should exhibit the properties of a weak electrolyte. In this case, the dependence of the molar conductivity on the concentration of sulfates of lithium, sodium and potassium in water-ethanol solutions will have a different appearance. Indeed, with an increase in the alcohol content in aqueous ethanol solutions, the molar conductivity decreases due to a decrease in the degree of electrolytic dissociation. With a decrease in the salt concentration in the solution, the molar electrical conductivity increases, approaching λ_0, for both water and water-alcohol solutions. As it follows from the obtained data, the dependence of λ on the total concentrations of Li_2SO_4, Na_2SO_4, K_2SO_4) for the water solution with a high correlation coefficient is transformed into a straight line in the coordinates of the Kolraush equation since the sulfates of lithium, sodium and potassium act as strong electrolytes in the aqueous medium. For water-ethanol solutions, a high correlation coefficient in the coordinates of the equation $4\lambda^3 \cdot C_0(M_2SO_4)^2 = K_{DIS} \cdot \lambda_0^3 - K_{DIS} - \lambda_0^2 \cdot \lambda$ is observed in the case of $[C_2H_5OH] > 50\%$ of the volume. This means that in these solutions lithium, sodium and potassium sulphates exhibit the properties of a weak electrolyte. When the alcohol content is from 10% to 40% by volume, sulfates of lithium, sodium and potassium are medium-strength electrolyte and, therefore, low correlation coefficients are characteristic of transformations in the coordinates of the equations Kohlrausch and $4\lambda^3 \cdot C_0(M_2SO_4)^2 = K_{DIS} \cdot \lambda_0^3 - K_{DIS} - \lambda_0^2 \cdot \lambda$.

Key words: aqueous ethanol, lithium sulphate, sodium sulphate, potassium sulphate, conductometry, electrolytic dissociation constants

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INTRODUCTION

Information about the electrochemical properties of aqueous organic solutions of alkali metal sulphates is extremely scarce. However, this information can serve as a theoretical and scientific basis for creating more optimal conditions for the processing and purification of inorganic salts. The authors [1, 2] showed a decrease in the electrical conductivity of sodium sulphate in aqueous acetone and aqueous dioxane and proposed mathematical equations describing the dependence of the conductivity on the salt concentration. The physicochemical properties of lithium, sodium, and potassium sulphates in aqueous solutions and melts were described in [3-6]. However, in the literature, there are practically no data on the physicochemical properties of alkali metal sulphates in aqueous-alcoholic solutions. The authors of [7-11] indicate the relevance and practical significance of studying the physicochemical properties of inorganic salts in aqueous-organic solutions. Earlier, in [12], we have shown that in aqueous-ethanol solutions the degree of electrolytic dissociation of sodium sulphate decreases with increasing alcohol concentration. In this case, Na_2SO_4 is converted from a strong electrolyte in an aqueous solution to an electrolyte of medium or low strength in aqueous ethanol solutions, depending on the alcohol content. This study was continued and the regularities of electrolytic dissociation of lithium, sodium and potassium sulphates in aqueous ethanol solutions were studied.

EXPERIMENTAL PART

The dietary ethanol used in the experiment was rectified, the residual water concentration was 4% by volume. Sulphate lithium of brand "C.P. (chemically pure) " sodium sulphate of brand "C.P." and potassium sulphate of brand "C.P." were additionally purified by recrystallization. A certain amount of ethyl alcohol was added to the initially prepared saturated aqueous solutions of lithium, sodium and potassium sulphates.

The resulting water-ethanol salt solutions were brought to a state of equilibrium by mixing with magnetic stirrers. The achievement of equilibrium state in the studied systems was checked by microcrystalline method [13]. The concentration of salts in saturated solutions was determined according to (R.S.S.) GOST [14], by titration with barium chloride in the presence of an indicator, dichromate. Specific electrical conductivity of sodium sulphate solutions æ (Ohm^{-1}-m^{-1}) was determined using a conductometer "HM-200". The molar conductivity λ ($Ohm^{-1}\cdot mol^{-1}\cdot m^2$) was found by the formula $\lambda = æ\cdot 1000/C_0(M_2SO_4)^2$, where $C_0(M_2SO_4)$ is the initial concentration of Li_2SO_4, Na_2SO_4, K_2SO_4.

During the main experiment, the original saturated solution was tenfold diluted using solvent (water was used as a solvent for aqueous solutions, and water-ethanol solutions were diluted by corresponding mixture of water with ethanol). The intrinsic electrical conductivity of the used solvents was determined experimentally. It was no more than 1% of the electrical conductivity of the solution.

RESULTS AND DISCUSSION

It is known [15, 16] that the solubility of lithium, sodium and potassium sulphates decreases with increasing alcohol content in a solution. The concentrations of salts in saturated water-ethanol solutions were determined by the method of volumetric titration described in [14] (Table 1).

Table 1

Molar concentrations of alkali metal sulphates in saturated aqueous ethanol solutions. T=25 °C

***Таблица 1.* Молярные концентрации сульфатов щелочных металлов в насыщенных водно-этанольных растворах. T=25 °C**

$[H_2O]:[C_2H_5OH]$, Volume fractions, %	$C_0(Li_2SO_4)$ Mol/L	$C_0(Na_2SO_4)$ Mol/L	$C_0(K_2SO_4)$ Mol/L
100:0	3.313	1.965	0.692
90:10	2.211	0.961	0.441
80:20	1.553	0.383	0.161
70:30	1.061	0.161	0.105
60:40	0.742	0.076	0.058
50:50	0.432	0.052	0.022
40:60	0.183	0.038	0.015
30:70	0.089	0.016	0.009
20:80	0.031	0.008	0.005
10:90	0.007	0.005	0.002

During the injection of alcohol not just the concentration but also the mass fraction of salt in the studied solutions decreases. In Table 2 the evidence of decreasing solubility of sulphate alkali metals in three-component systems: Li_2SO_4-H_2O-C_2H_5OH, Na_2SO_4-H_2O-C_2H_5OH and K_2SO_4-H_2O-C_2H_5OH at 25 °C both described in the literature [15, 16] and obtained by us additionally in the field of low salt concentrations in the system is given.

Table 2

The solubility of sulfates of lithium, sodium and potassium in water-ethanol solutions. T= 25 °C

***Таблица 2.* Растворимость сульфатов лития, натрия и калия в водно-этанольных растворах. T= 25 °C**

The composition of a saturated solution, wt.%								
System Li_2SO_4-H_2O-C_2H_5OH			System Na_2SO_4-H_2O-C_2H_5OH			System K_2SO_4-H_2O-C_2H_5OH		
Li_2SO_4	H_2O	C_2H_5OH	Na_2SO_4	H_2O	C_2H_5OH	K_2SO_4	H_2O	C_2H_5OH
25.60	74.40	0.00	21.90	78.10	0.00	11.10	88.90	0.00
19.80	73.65	6.55	12.22	80.62	7.16	7.25	84.40	7.50
15.08	70.77	14.15	5.32	78.90	15.78	2.84	80.97	16.19
11.04	66.25	22.71	2.36	72.71	24.93	1.80	73.10	25.10
8.13	59.91	31.96	1.16	64.46	34.38	1.10	64.50	34.40
4.99	52.79	42.22	0.81	55.10	44.08	0.42	55.28	44.30
2.24	44.43	53.33	0.61	45.18	54.21	0.31	45.29	54.40
1.13	34.49	64.38	0.26	34.80	64.94	0.18	34.82	65.00
0.41	23.71	75.88	0.14	23.78	76.09	0.11	23.78	76.10
0.09	12.18	87.72	0.09	12.18	87.73	0.04	12.16	87.76

It can be seen from the Table 2 that with the increase in alcohol concentration the solubility of lithium, sodium and potassium sulphates decreases more than two-fold. In our opinion, it is caused by the change in the dielectric constant of the liquid phase. Water (ε = 78.53) and ethanol (ε = 24.30) differ sufficiently in dielectric permittivity. In that way, it is possible to prepare the solvent with different dielectric permittivity by changing the alcohol content in a mixture with water, thus, affecting the equilibrium state of the salt in solution. When calculating the Table 2 data, the experimentally determined values of the density of saturated solutions of lithium, sodium and potassium sulphates in aqueous ethanol solutions were used (Table 3).

The decrease in the density of solutions is caused not only by the addition of alcohol, which has a lower density compared to water, but also by a decrease in the concentration of dissolved salt.

The characteristic of electrolytic dissociation of lithium, sodium and potassium sulphates were studied by conductometric method, same as in [12], by finding the dependence of molar conductivity on the salt concentration in the solution. The concentration of Li_2SO_4, Na_2SO_4, K_2SO_4 was varied by diluting the saturated solutions with an appropriate solvent (water or aqueous ethanol). In aqueous solutions, the sulphates of lithium, sodium and potassium are strong electrolytes and, therefore, the interdependence of molar conductivity and salt concentration obeys the equation Kohlrausch [17-21].

$$\lambda = \lambda_0 – a\cdot C_0(M_2SO_4)^{½} \qquad (1)$$

where λ is the molar electrical conductivity of the solution; λ_0 is the maximum molar electrical conductivity of the solution; $C_0(M_2SO_4)$ is the initial concentration of lithium, sodium or potassium sulphate; a is the parameter characterizing the electrophoretic and relaxation effects of ion inhibition.

At low concentrations of ethanol (10-40% vol.) in a solution Li_2SO_4, Na_2SO_4, K_2SO_4 exhibit the properties of an electrolyte of medium strength [12].

Table 3.
The density of saturated solutions of lithium, sodium and potassium sulfates in water-ethanol solutions. T= 25 °C
Таблица 3. **Плотность насыщенных растворов сульфатов лития, натрия и калия в водно-этанольных растворах. T= 25 °C**

$[H_2O]:[C_2H_5OH]$, Volume fractions in%	ρ ($H_2O + C_2H_5OH$) kg/m^3	ρ ($H_2O + C_2H_5OH + M_2SO_4$) kg/m^3		
		Li_2SO_4	Na_2SO_4	K_2SO_4
100:0	997.05*	1245.01	1240.02	1093.01
90:10	984.71	1150.04	1175.04	1035.05
80:20	973.62	1095.02	1100.03	990.06
70:30	962.21	1040.01	1000.01	966.01
60:40	948.01	1000.04	950.04	950.04
50:50	930.23	970.03	940.03	911.03
40:60	909.11	950.01	920.01	901.07
30:70	885.52	902.04	885.02	878.09
20:80	859.34	890.02	865.04	858.04
10:90	829.21	860.01	845.03	839.01

* The data from [17] was used, the density of the solvent (mixture of water with ethanol) was found experimentally

* Использованы данные из [17], плотность растворителя найдена экспериментально (смесь воды с этанолом)

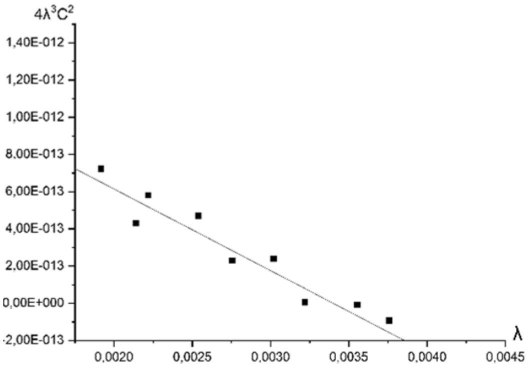

Fig. The linear dependence of molar conductivity on the concentration of lithium sulfate in the coordinates of equation (2). $[H_2O]$: $[C_2H_5OH]$= 10:90, volume percentages. The correlation coefficient is 0.9999

Рис. Линейная корреляция зависимости молярной электропроводности от концентрации сульфата лития в координатах уравнения (2). $[H_2O]$: $[C_2H_5OH]$ = 10:90 объемные доли в %. Коэффициент корреляции равен 0,9999

At a certain concentration of alcohol in the solution (> 50% vol.), these salts become the weak electrolytes and, therefore, they are characterized by the following equation [12]:

$$4\lambda^3\cdot C_0(M_2SO_4)^2 = K_{DIS}\cdot\lambda_0^3 - K_{DIS}\,\lambda_0^2\cdot\lambda \qquad (2)$$

From the linear graphic dependence $4\lambda^3\cdot C_0(M_2SO_4)^2$ on λ (Figure) the slope tangent, which is equal to $K_{DIS}\,\lambda_0^2$, and the intercept (which is equal to $K_{DIS}\cdot\lambda_0^3$), were determined.

These values were used to determine

$$\lambda_0 = K_{DIS}\cdot\lambda_0^3/K_{DIS}\cdot\lambda_0^2 \qquad (3)$$

and salt dissociation constants in aqueous ethanol solution

$$K_{DIS} = \text{slope angle tangent}/\lambda_0^2 \qquad (4)$$

The obtained values of the electrolytic dissociation constants of the studied salts are summarized in the Table 4.

Therefore, as we can see from the Table 4, the values of the dissociation constant of alkali metal sulphates with increasing alcohol concentration decreases. This is due to the decrease in the polarity of the liquid phase during the transition from a strongly polar solvent (water) to a mixture with less polar (ethanol). The data of Table 4 also clearly indicate the influence of the nature of the salt on the value of the electrolytic dissociation constant: with an increase in the

Table 4

Values of electrolytic dissociation constants of lithium, sodium and potassium sulphates in aqueous ethanol solutions T = 25 °C

***Таблица 4*. Значения констант электролитической диссоциации сульфатов лития, натрия и калия в водно-этанольных растворах. Т = 25 °С**

Volume fraction of ethanol, % vol.	K_{DIS} (mol/l)^2		
	Li_2SO_4	Na_2SO_4	K_2SO_4
50	$(3.54\pm0.01)\cdot10^{-3}$	$(1.95\pm0.01)\cdot10^{-3}$	$(2.42\pm0.01)\cdot10^{-6}$
60	$(1.01\pm0.31)\cdot10^{-3}$	$(5.97\pm0.01)\cdot10^{-4}$	$(1.25\pm0.06)\cdot10^{-6}$
70	$(6.54\pm0.21)\cdot10^{-4}$	$(2.29\pm0.05)\cdot10^{-4}$	$(8.96\pm0.02)\cdot10^{-7}$
80	$(1.08\pm0.01)\cdot10^{-4}$	$(2.31\pm0.01)\cdot10^{-5}$	$(6.20\pm0.40)\cdot10^{-7}$
90	$(3.80\pm0.03)\cdot10^{-5}$	$(1.77\pm0.02)\cdot10^{-5}$	$(1.06\pm0.01)\cdot10^{-7}$

size of the cation, the electrolytic dissociation constant decreases. The total effect of these factors seems to reduce the polarity of the oxygen-metal bond, which reduces K_{DIS}.

Using the more sensitive conductometer and finding the salt concentration in the solution by volume titration instead of the gravimetric one allowed us to clarify the values of the electrolytic dissociation constants Na_2SO_4, which were previously given in [12].

The data obtained as a reference data can be used as a theoretical and scientific basis for cleaning and obtaining inorganic substances [12]. For the first time, concentrations of lithium, sodium, and potassium sulphates in water-ethanol solutions with different alcohol contents were determined using the bulk titration method.

In Table 4 data, two regularities of electrolytic dissociation of salts are observed. First, a decrease in the dissociation constants of alkali metal sulphates with an increase in the volume fraction of ethanol indicates that alcohol additives significantly reduce the degree of electrolytic dissociation of salts. Second, the constant, hence the degree, of electrolytic dissociation depends on the nature of the cation and decreases in the row Li-Na-K.

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