Temperature and Concentration Dependencies of the Electrical Conductance of Potassium Tellurite Solutions in Ordinary and Heavy Water

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On the basis of conductometric measurements, empirical equations were derived, describing the temperature and concentration dependencies of the electrical conductance of K_2 TeO₃ solutions in ordinary and heavy water. The values of the equivalent conductivity of the ions at infinite dilution in both solvents were determined in the temperature region (12 to 45) °C, using the expressions for 1:2 valent electrolytes of the semiempirical Fuoss– Onsager equation. These values were compared with respect to the nature of the solvent.

Introduction

Tellurites, in particular K₂TeO₃, are not studied enough. There are some papers concerning the basic thermodynamic functions of the state of K₂TeO₃,^{1,2} its structure,³⁻⁵ IR spectrum,⁶ and thermal stability.^{7,8} The solid K₂TeO₃ is used as a component in the production of tellurite glasses, 9^{-13} and the aqueous solutions of K₂TeO₃ are suitable for producing marginally soluble tellurites from which the corresponding tellurides are prepared by reduction with H₂ or CO under heating. Some of these tellurides have interesting semiconducting properties.^{14–17} K₂TeO₃ itself has a ferroelectric behavior.¹⁸ Recently, K₂TeO₃ and its solutions have been used in medicine because they have an anti-sickling effect,^{19,20} and in microbiology the oxyanion tellurite, TeO_3^2 -, can be used as a tool to detect and quantify the release in soil microcosms of Pseudomonas pseudoalcaligenes KF707²¹ and as a reagent for rapid drug susceptibility testing of Escherichia coli.²²⁻²⁸

The use of the solvents H₂O and D₂O, which have different densities, viscosities, and dielectric constants, allows the solvent isotope effect to be studied using the electrical conductance.²⁹⁻³² According to published results,^{33–36} the electrical conductance of ions is always greater in light water than in heavy water. For example, at 25 °C the differences between the limiting equivalent conductance at infinite dilution of OH⁻ and OD⁻ as well as H⁺ and D⁺ are 41.4 and 33.3 %, respectively.^{35,36} For other ions this difference is between (15 and 19) %.33,34 With increasing temperature these differences decrease. It is the first evidence for the distinctive differences in the behavior of an isotopically substituted substance. In going from light to heavy water, one of the bulk properties upon which the conductance depends, the viscosity, changes by 23 %, while another bulk property, the dielectric constant, changes by only 0.4 %.³³ The practical implementation of some heavy water solutions is connected with their use in the control of nuclear activity in CANDU nuclear reactors^{37,38} as well as for studies of the socalled "cold fusion" phenomenon.39,40

All the facts, mentioned above, have led to an extensive research of the electrical conductivity of diluted aqueous solutions of $K_2 TeO_3$ in a broad temperature range using isotopically substituted solvents. The present paper reports the

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temperature and concentration dependencies of the electrical conductivity of potassium tellurite solutions in ordinary and heavy water. The coefficients in the corresponding empirical equations are derived, and the influence of the isotopic substitution in the solvent on the electrical conductance of $K_2 TeO_3$ solutions is compared.

Experimental Section

The initial solutions were prepared by diluting a precisely weighed amount of K2TeO3 (Aldrich) in double-distilled ordinary water with specific conductivity $\kappa = 9.2 \pm 10^{-7}$ S·cm⁻¹ at 25 °C and in 99.85 % D₂O (Fluka) with specific conductivity $\kappa = 3.8 \pm 10^{-7}$ S·cm⁻¹ at 25 °C, which were placed in a 250 cm3 volumetric flask at 20 °C. The concentration of Te(IV) in the initial solutions was determinated by oxidation with a solution of K₂Cr₂O₇, an excess of which was titrated against a standardized solution of ammonium iron(II) sulfate solution (Mohr's salt).⁴¹ A series of eight solutions with concentrations from (0.1000 to 0.0028) equiv-dm⁻³ was prepared by diluting an aliquot of the initial solutions with the appropriate solvent in a 100 cm³ volumetric flask, placed in a thermostat at 20 °C. The specific conductivities of these solutions were measured using a digital conductivity meter, Inolab Level-1 WTW (Germany), having a conductometric cell constant 0.4752 \pm 0.0002 cm⁻¹. According to the technical specification of the apparatus, the relative error of the electrical conductivity measurements should be \pm 0.02 %. The measurements were carried out at 1° increments in the temperature range (5 to 45) °C for light water solutions and in the temperature interval (12 to 45) °C for heavy water solutions. The temperature was maintained with a precision of \pm 0.05 °C using an Ultrathermostat U-1; 50 cm³ of each sample was pipetted in a 70 cm³ glass container equipped with a water jacket. The measurements were performed under constant pressure and continuous stirring (600 rpm) by an electromagnetic stirrer. To avoid changes in the electrical conductivity of the sample due to dissolution of CO_2 or other gases, present in air, the container was sealed with a rubber cap through which the conductivity cell had been inserted. The relative error of the measurements (temperature, concentration and specific conductivity) was < 0.1 % during the experiments. The experimental data were processed by the mean square method.



Figure 1. Temperature and concentration dependence of the specific conductivity of K_2TeO_3 in H_2O (1) and D_2O (2).

Results and Discussion

Figure 1 shows the concentration and temperature dependence of the specific conductivity of K₂TeO₃ in ordinary and heavy water, respectively. As can be seen from the figure, the specific conductivity of K₂TeO₃ solutions in H₂O was higher than that in D₂O at any concentration and temperature. This is due to the higher density and viscosity of D₂O as compared to H₂O. Using the mathematical approach described in detail earlier,⁴² the coefficients in the empirical equations, describing the dependence $\kappa = f(t, c)$ of the studied solutions, were found to be:

$$\begin{aligned} \kappa_{\rm H_2O} &= 3.6489 \times 10^{-5} + 1.3175 \times 10^{-6}t + 2.5872 \times 10^{-8}t^2 + \\ &(5.4472 \times 10^{-2} + 1.7834 \times 10^{-3}t + 6.0974 \times 10^{-6}t^2)c - \\ &(5.4571 \times 10^{-2} + 2.0140 \times 10^{-3}t + 1.4208 \times 10^{-5}t^2)c^2 \ (1) \\ &\kappa_{\rm D_2O} &= 1.1869 \times 10^{-5} + 5.7919 \times 10^{-7}t + 5.3992 \times 10^{-9}t^2 + \\ &(4.4066 \times 10^{-2} + 1.6984 \times 10^{-3}t + 4.9961 \times 10^{-6}t^2)c - \\ &(4.7497 \times 10^{-2} + 2.2695 \times 10^{-3}t + 4.9045 \times 10^{-6}t^2)c^2 \ (2) \end{aligned}$$

were *t* is the temperature in °C and *c* is the analytical concentration of the solutions in equiv•dm⁻³. These empirical equations describe the surfaces presented in Figure 1 and can be used to calculate the analytical concentration of potassium tellurite in light or heavy water. The specific conductance of the solutions, calculated according to eqs 1 or 2, was determined with a relative error about 0.8 % only for the highest concentrations and temperatures of the solutions. In all other cases the relative error was lower than 0.2 %. The standard deviations were between 0.0071 and 0.0249. The Student's criterion was calculated at $\alpha = 0.95$ and had values between 2.128 and 2.306.

The values of the equivalent conductivity Λ of K₂TeO₃ solutions in H₂O and D₂O were calculated, using the values of the specific conductivity κ of the solutions at corresponding temperatures and concentrations and the relationship between the specific and the equivalent conductivities (Λ) of the solutions:⁴³

$$\Lambda = \frac{\kappa}{c} = \sum_{i=1}^{n} \frac{|z_i| c_i \lambda_i}{c}$$
(3)

where λ_i are the ionic conductances, c_i and z_i are concentrations and charges of individual ions present in the solution and *c*



Figure 2. Temperature and concentration dependence of Λ for K₂TeO₃ in H₂O (1) and in D₂O (2).

Table 1. Values of the Equivalent Conductance $\Lambda/S \cdot cm^2 \cdot equiv^{-1}$ of $K_2 TeO_3$ in H_2O and D_2O at Different Concentrations and Temperatures

С	t/°C							
equiv•dm ⁻³	12	15	20	25	30	35	40	45
K ₂ TeO ₃ -H ₂ O								
0.09837	69.33	74.62	83.56	92.61	102.06	111.92	121.89	132.05
0.04919	73.39	79.08	88.64	98.39	108.76	119.33	130.11	141.29
0.03935	74.97	80.56	90.47	100.38	111.05	121.73	132.91	144.35
0.02951	76.58	82.34	92.51	103.02	113.86	125.04	136.56	148.42
0.01967	78.80	84.95	95.53	106.51	118.20	129.89	141.84	154.30
0.00984	82.37	88.92	100.15	111.84	124.03	136.84	150.20	163.77
0.00590	84.75	91.53	103.22	115.25	127.97	141.27	155.25	169.32
0.00393	87.02	93.89	105.85	118.45	131.55	145.29	159.41	174.05
KaTeOa-DaO								
0.09007	58.68	63.45	71.78	80.27	89.15	98.09	107.42	116.80
0.04504	61.94	67.16	76.04	85.04	94.36	104.02	113.90	123.78
0.03603	63.14	68.42	77.44	86.73	96.17	105.88	116.01	126.14
0.02702	64.03	69.39	78.65	88.27	98.08	108.07	118.06	128.61
0.01802	65.26	70.81	80.30	90.12	100.22	110.49	120.98	132.08
0.00901	67.20	72.81	82.57	92.51	102.89	113.54	124.58	135.74
0.00540	68.15	73.80	83.70	93.80	104.26	114.81	126.02	137.13
0.00360	68.61	74.17	84.17	94.44	104.86	115.56	126.67	138.06

denotes the analytical electrolyte concentration. Some of the results are presented in Table 1.

Figure 2 shows the dependence $\Lambda = f(t, c^{1/2})$ for the solutions of K₂TeO₃ in H₂O and D₂O, respectively, which is based on the results for the equivalent conductance at different concentrations and temperatures. The values of the limiting equivalent conductance Λ° of K₂TeO₃ solutions in H₂O and D₂O at the corresponding temperatures can be obtained according to the Kohlrausch equation:⁴⁴

$$\Lambda = \Lambda^{\rm o} - A\sqrt{c} \tag{4}$$

which describes a linear dependence of the equivalent conductivity of strong electrolytes on the square root of solution concentration. The extrapolation to a zero concentration gives the limiting equivalent conductivity Λ° , and the slope of the straight line—the *A* coefficient.

The coefficient A in eq 4 can be theoretically calculated using the limiting Onsager law:⁴⁴

$$\Lambda = \Lambda^{\circ} - (B_1 \Lambda^{\circ} + B_2) \sqrt{c} = \Lambda^{\circ} - S \sqrt{c}$$
(5)

where the coefficients B_1 and B_2 take into account relaxation effects of the ions motion in the solution and electrophoretic

effects, respectively. These coefficients depend on the absolute temperature *T*, viscosity η , and dielectric constant ϵ of the solvent.

The values of Λ° obtained from eq 4 at different temperatures were used in the semiempirical Fuoss–Onsager equation^{42,45–48} to calculate more precisely the values of Λ° :

$$\Lambda = \Lambda^{o} - S\sqrt{c} + Ec \ln c + J_{1}c + J_{2}c^{3/2}$$
(6)

Equation 6 is applicable for non-associated ionophores. Here, *S* is the limiting slope of the Onsager equation and the *E*, J_1 , and J_2 values are determined by relaxation and electrophoretic effects and depend on Λ° , η , ϵ , and *T*. Besides, the coefficients J_1 and J_2 depend on the distances of the closest approach a° between cation and anion centers. The coefficients of eq 6 for 1:2 valent electrolytes are described in detail by Barthel⁴⁹ and Vlaev et al.:⁵⁰

$$\begin{split} \Lambda &= \Lambda^{\circ} - \left\{ \frac{6.4671 \times 10^{6} \Lambda^{\circ}}{(\epsilon T)^{3/2} [1 + \lambda_{+}^{\circ} / \Lambda^{\circ} + 0.8165 (1 + \lambda_{+}^{\circ} / \Lambda^{\circ})^{1/2}]} + \right. \\ \frac{214.3533}{\eta(\epsilon T)^{1/2}} \left\} c^{1/2} + \left[\frac{3.5289 \times 10^{13} \Lambda^{\circ}}{(\epsilon T)^{3}} - \frac{7.7959 \times 10^{8}}{\eta(\epsilon T)^{2}} \right] c \ln c + \\ &\left. \left\{ \left\{ \frac{7.0579 \times 10^{13}}{(\epsilon T)^{3}} \right[- 2.68 \times 10^{7} (\mathring{a} \epsilon T)^{3} + 1.7911 \times \right. \\ 10^{5} (\mathring{a} \epsilon T)^{2} + 5.985 \times 10^{2} \mathring{a} \epsilon T + 0.9074 + \ln \frac{8.7091 \times 10^{9} \mathring{a}}{(\epsilon T)^{1/2}} \right] \right\} \end{split}$$

$$\Lambda^{o} + \frac{214.3533}{\eta(\epsilon T)^{1/2}} \left[\frac{6.4671 \times 10^{6} \Lambda^{o}}{(\epsilon T)^{3/2} [1 + \lambda_{+}^{o} / \Lambda^{o} + 0.8165(1 + \lambda_{+}^{o} / \Lambda^{o})^{1/2}]} + \frac{7.9832 \times 10^{9} \mathring{a}}{(\epsilon T)^{1/2}} - \frac{3.6377 \times 10^{6}}{(\epsilon T)^{3/2}} \left(1.017 + \ln \frac{8.7091 \times 10^{9} \mathring{a}}{(\epsilon T)^{1/2}} \right) \right] \right\}$$

$$c + \left\{ \left\{ \frac{1.4524 \times 10^{21}}{(\epsilon T)^{9/2}} [2.68 \times 10^{7} (\mathring{a} \epsilon T)^{3} + 1.7912 \times 10^{5} (\mathring{a} \epsilon T)^{2} - 3.225 \times 10^{2} \mathring{a} \epsilon T] \right\} \Lambda^{o} + \frac{1.6258 \times 10^{22} \mathring{a}^{2}}{\eta(\epsilon T)^{3/2}} \right\} c^{3/2} (7)$$

For the temperature dependence of the dielectric constant ϵ and the viscosity η of ordinary water, the following empirical equations were used:^{51–53}

$$\log \epsilon_{\rm H_2O} = 1.94409 - 1.991 \times 10^{-3} t \tag{8}$$

$$\log \frac{\eta_t}{\eta_{20}} = \frac{1.1709(20-t) - 0.001827(t-20)^2}{t+89.93}$$
(9)

For heavy water, eqs 10 and 11 were used:^{51,54}

$$\log \epsilon_{\rm D,0} = 1.94275 - 2.013 \times 10^{-3} t \tag{10}$$

$$\log \frac{\eta_t}{\eta_{20}} = \frac{1.3580(20-t) - 0.00067(t-20)^2}{t+96.71}$$
(11)

The equation used for the temperature dependence of the limiting equivalent conductivity of K^+ ions in ordinary water was^{55}

$$\lambda_{\rm K^+}^{\rm o}({\rm H_2O}) = 40.5017 + 1.2194t + 4.1859 \times 10^{-3}t^2 \quad (12)$$

and in heavy water:56

$$\lambda^{\rm o}_{\rm K^+}({\rm D}_2{\rm O}) = 32.3892 + 1.1895t + 2.0463 \times 10^{-3}t^2 \quad (13)$$

Table 2. Values of the Limiting Equivalent Conductance $\Lambda^{\circ/}$ S·cm²·equiv⁻¹ and the Coefficients *S*, *E*, *J*₁, *J*₂ and a in Equation 7 for K₂TeO₃ Solutions in H₂O and in D₂O at Different Temperatures

t	parameter								
°C	Λ^{o}	S	Ε	J_1	J_2	å∕Å	$\sigma(\Lambda)$		
	K ₂ TeO ₃ -H ₂ O								
5	84.40	148.74	128.31	572.91	-157.94	1.771	0.0476		
10	97.28	173.36	149.81	666.60	-186.15	1.760	0.0541		
12	102.56	183.63	158.77	706.21	-198.05	1.757	0.0576		
15	110.81	199.66	173.14	768.03	-217.20	1.751	0.0611		
20	125.19	227.81	199.02	877.19	-252.02	1.740	0.0687		
25	140.21	257.64	227.15	991.72	-290.37	1.725	0.0757		
30	156.08	289.33	258.29	1117.52	-333.13	1.714	0.0850		
35	172.58	322.70	292.10	1251.71	-380.06	1.703	0.0912		
40	189.82	357.88	329.17	1395.62	-431.93	1.692	0.0984		
45	207.62	394.72	369.20	1550.21	-488.54	1.683	0.1079		
K ₂ TeO ₃ -D ₂ O									
12	81.95	145.03	129.94	619.74	-159.96	1.921	0.0631		
15	88.93	158.72	142.04	678.96	-176.02	1.919	0.0708		
20	101.17	182.96	163.97	783.38	-205.67	1.911	0.0814		
25	113.82	208.63	187.35	895.04	-237.91	1.904	0.0933		
30	126.87	235.73	212.36	1015.50	-272.99	1.900	0.1054		
35	140.32	264.21	239.17	1143.98	-311.32	1.896	0.1193		
40	154.30	294.20	268.45	1281.59	-353.90	1.891	0.1316		
45	168.70	325.57	300.05	1427.66	-400.67	1.885	0.1466		

Substituting the values of ϵ , η , $\Lambda_{1/2K_2TeO_3}^o$, and $\lambda_{K^+}^o$ for the corresponding temperature and solvent into eq 7, the coefficients *S*, *E*, *J*₁, and *J*₂ were calculated, which allowed the temperature and concentration dependencies of the equivalent conductivity of K₂TeO₃ in ordinary and heavy water, respectively, to be studied. The "goodness" of the fit of the determined equivalent conductance is expressed by the mean-square deviation $\sigma(\Lambda)$, calculated by the equation:

$$\sigma(\Lambda) = \sqrt{\frac{\sum_{i=1}^{n} \left(\Lambda_{i,\exp} - \Lambda_{i,\text{calc}}\right)^{2}}{n-1}}$$
(14)

where n is the number of conductance-concentration pairs.

The obtained values of $\Lambda_{1/2K_{1}TeO_{3}}^{o}$ as well as the mean-square deviation $\sigma(\Lambda)$ are presented in Table 2. It can be seen from Table 2 that the values of Λ^{o} increase with temperature, while those for K₂TeO₃ in H₂O remain higher due to the lower density and viscosity of ordinary water as compared to heavy water. The values of the limiting equivalent conductivity Λ^{o} of K₂TeO₃ in H₂O and in D₂O in the temperature range (12 to 45) °C can be calculated by the following empirical equations:

$$\Lambda^{o}_{1/2K_{2}TeO_{3}}(H_{2}O) = 72.1555 + 2.3632t + 1.4442 \times 10^{-2}t^{2}$$
(15)

$$\Lambda^{0}_{1/2K_{2}TeO_{3}}(D_{2}O) = 55.0817 + 2.1262t + 8.8621 \times 10^{-3}t^{2}$$
(16)

The values of the limiting equivalent conductivity λ^{o} for TeO_{3}^{2-} anions in H₂O and in D₂O, respectively, can be calculated in the studied temperature range knowing the temperature dependence of the limiting equivalent conductivity of K⁺ ions in both ordinary and heavy water, which are presented in Table 3.

Table 3 shows that the values of $\Lambda_{1/2K_2TeO_3}^{o}$, $\lambda_{K^+}^{o}$, and $\lambda_{1/2TeO_3^{2-}}^{o}$ in H₂O are higher than the same values in D₂O. This can be attributed to the lower values of the density and viscosity of H₂O as compared to D₂O. It can be also seen that, at a given temperature, the values of the Walden's product for both solvents are practically the same, slightly decreasing with an

Table 3. Values of the Limiting Equivalent Conductance at Infinite Dilution of the Molecule and the Ions of K₂TeO₃: $\Lambda^{o}_{1/2K_{2}TeO_{3}}$, $\lambda^{o}_{K^{+}}$, $\lambda^{o}_{1/2TeO_{3}^{2\rightarrow}}$ and Walden's Product ($\Lambda^{\circ}\eta$) for Its Ordinary and Heavy Water Solutions at Certain Temperatures

t	K ₂ TeO ₃ -H ₂ O				K ₂ TeO ₃ -D ₂ O			
°C	$\Lambda^o_{1/2K_2TeO_3}$	$\Lambda^{\circ}\eta$	$\lambda^o_{K^+}$	$\lambda^o_{1/2TeO_3{}^{2-}}$	$\Lambda^o_{1/2K_2TeO_3}$	$\Lambda^{\circ}\eta$	$\lambda^{\rm o}_{K^+}$	$\lambda^o_{1/2TeO_3^{2-}}$
12	102.56	1.27	55.74	46.82	81.95	1.29	46.96	34.99
15	110.81	1.26	59.73	51.07	88.93	1.28	50.69	38.24
20	125.19	1.25	66.56	58.62	101.17	1.26	57.00	44.17
25	140.21	1.25	73.60	66.61	113.82	1.25	63.41	50.41
30	156.08	1.24	80.85	75.23	126.87	1.23	69.92	56.96
35	172.58	1.24	88.31	84.27	140.32	1.22	76.53	63.79
40	189.82	1.24	95.98	93.85	154.30	1.21	83.24	71.06
45	207.62	1.24	103.85	103.77	168.70	1.20	90.06	78.64

increase of temperature. The changes of the isotopic ratios $\Lambda^o_{1/2K_2TeO_3}~(H_2O)/\Lambda^o_{1/2K_2TeO_3}~(D_2O),~\lambda^o_{K^+}~(H_2O)/\lambda^o_{K^+}~(D_2O),$ and $\lambda^o_{1/2TeO_3^{2-}}(H_2O)/\lambda^o_{1/2TeO_3^{2-}}(D_2O)$ with an increase of temperature can be assessed using Figure 3.



Figure 3. Temperature dependence of the isotopic ratios: $\Lambda_{1/2K_2TeO_3}^{o}(H_2O)/\Lambda_{1/2K_2TeO_3}^{o}(D_2O)$ (1); $\lambda_{K^+}^{o}(H_2O)/\lambda_{K^+}^{o}(D_2O)$ (2); and $\lambda_{1/2TeO_3^{2-}}^{o}(H_2O)/\lambda_{1/2TeO_3^{2-}}^{o}(H_2O)/\lambda_{1/2TeO_3^{2-}}^{o}(D_2O)$ (3).

Figure 3 shows that the values of the isotopic ratios slightly decrease with an increase of temperature. A similar tendency was observed for solutions of other salts in light and heavy water,^{34,35} which could be explained by a decrease of the viscosities of ordinary and heavy water with an increase of temperature.

Conclusion

For the first time the temperature and concentration dependence of the specific and equivalent conductivity of K₂TeO₃ is studied in light and heavy water solutions. The coefficients of the corresponding empirical equations are calculated. On this basis, the values of the limiting equivalent conductance of TeO_3^{2-} ions are determined in both solvents in the temperature region (12 to 45) °C. The temperature dependence is determined for the isotopic ratios $\Lambda^{\circ}_{1/2K_2TeO_3}$ (H₂O)/ $\Lambda^{\circ}_{1/2K_2TeO_3}$ (D₂O), $\lambda^{\circ}_{K^+}$ (H₂O)/ $\lambda^{\circ}_{K^+}$ (D₂O), and $\lambda^{\circ}_{1/2TeO_3^{2-}}$ (H₂O)/ $\lambda^{\circ}_{1/2TeO_3^{2-}}$ (D₂O). These isotopic ratios decrease with an increase of temperature. Since the comparisons of the electrical conductivity of ordinary and heavy water solutions are insufficient in the literature, the implemented study and the presented results add to the knowledge in this field of study. It may be concluded that the conductometric method can give useful information about the solution structure, the state of the ions, and the influence of the solvent isotopic effect on their properties. As the kinetic isotope effect can be observed not only when reagents consist of different isotopes but also when the used solvents are isotope substituted by H₂O and D₂O, it is necessary that the values of the specific and the equivalent coductivity are compared with respect to the solvent-ordinary and heavy water. On this basis, the role of the solvent for the dissociation of the dissolving substance, the ion solvation, and the kinetics of ionic reactions in solutions can be estimated.

Note Added after ASAP Publication. The wrong concentrations were included in Table 1 in the version published ASAP January 6, 2007; the corrected version was published ASAP February 12, 2007.

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