Conductance Studies of Aqueous Succinic Acid

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Conductance measurements of aqueous solutions of succinic acid and of di-sodium succinate were performed from 278.15 to 308.15 K and the limiting conductances $\lambda^0(1/2\ \mathrm{Succ}^{2-})$ are reported. The Walden product is independent of temperature: $\lambda^0(1/2\ \mathrm{Succ}^{2-}) \cdot \eta(T) = 0.503 \pm 0.001$. The salt conductances closely obey the Onsager limiting law. The evaluation of the equilibrium constants for the primary and secondary steps of dissociation, K_1 and K_2, and the limiting conductances of the hydrosuccinate ion, $\lambda^0(\mathrm{HSucc}^-)$, are discussed using the Quint and Viallard conductance equation.

1. Introduction

This work is a continuation of our studies on aqueous solutions of polybasic organic acids [1], where it is difficult to obtain a simple interpretation of conductance measurements due to the overlapping dissociation equilibria. From many reported conductances of organic acids only in few cases the results for dibasic acids are analysed [2–4], and even then the contribution coming from the second dissociation step is usually considered as a small correction to the measured conductance. Succinic acid, a weak unsymmetrical 1:2 electrolyte is considered here in a similar way as citric acid [1] by using the Quint and Viallard conductance equation.

The conductances of aqueous succinic acid solutions have already been measured in 1912 by Jones [5]. Neutral and acidic succinates were considered by Jeffery and Vogel [6], Davies [2] and Topp and Davies [7]. Dissociation constants of succinic acid at room temperature were determined many times, mainly by potentiometric methods [8, 9]. Pinching and Bates [10, 11] performed a systematic evaluation of the dissociation constants from 273.15 to 323.15 K.

2. Experimental

The Fluka puriss. p.a. reagents anhydrous succinic acid (>99.5%), and di-sodium succinate (>98%), were used without further purification.

The solutions were prepared by weight, and conversion from molalities to molarities was performed with the help of the density of pure water at given temperature for dilute solutions ($c < 1.1 \cdot 10^{-3}$ mol dm^{-3}) of succinic acid and di-sodium succinate. Water with a specific conductance of less than $2 \cdot 10^{-7}$ S cm^{-1} was used for the solvent corrections of the specific conductances.

A detailed description of the measuring system and applied procedures is given in [1, 12, 13].

3. Data Analysis

The properties of dilute aqueous solutions of succinic acid are usually interpreted in terms of the successive dissociation steps:

$$\mathrm{H_2Succ} \rightarrow \mathrm{H^+} + \mathrm{HSucc^-}; \quad K_1, \tag{1a}$$

$$\mathrm{HSucc^-} \rightarrow \mathrm{H^+} + \mathrm{Succ^{2-}}; \quad K_2. \tag{1b}$$

The equilibrium constants of these reactions are

$$K_1 = \frac{[\mathrm{H^+}]\,[\mathrm{HSucc^-}]\,F_1}{[\mathrm{H_2Succ}]}, \tag{2a}$$

$$K_2 = \frac{[\mathrm{H^+}]\,[\mathrm{Succ^{2-}}]\,F_2}{[\mathrm{HSucc^-}]}, \tag{2b}$$

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where F_i denotes corresponding quotients of the activity coefficients:

$$F_1 = \frac{f_{\mathrm{H^+}}\, f_{\mathrm{HSucc^-}}}{f_{\mathrm{H_2Succ}}}, \tag{3a}$$

$$F_2 = \frac{f_{\mathrm{H^+}}\, f_{\mathrm{Succ^{2-}}}}{f_{\mathrm{HSucc^-}}}. \tag{3b}$$

In terms of the total degree of dissociation α and the degrees associated with the primary and secondary steps of dissociation, α_1 and α_2, the concentrations of species present in the solution are

$$[\mathrm{H^+}] = c(\alpha_1 + 2\alpha_2), \tag{4a}$$

$$[\mathrm{H_2Succ}] = c(1-\alpha), \tag{4b}$$

$$[\mathrm{HSucc^-}] = c\,\alpha_1, \tag{4c}$$

$$[\mathrm{Succ^{2-}}] = c\,\alpha_2, \tag{4d}$$

where c is the total (analytical) concentration of succinic acid and

$$\alpha = \alpha_1 + \alpha_2\,. \tag{5}$$

Use of (2) and (4) yields

$$K_1 = \frac{c(\alpha_1 + 2\alpha_2)\,\alpha_1}{1-\alpha} F_1\,, \tag{6a}$$

$$K_2 = \frac{c(\alpha_1 + 2\alpha_2)\,\alpha_2}{\alpha_1} F_2\,. \tag{6b}$$

For given values of the equilibrium constants and activity coefficients it follows at concentration c, that the degrees of dissociation α_1 and α_2 can successively be evaluated by an iterative solution of two quadratic equations:

$$\alpha_1 = \frac{1}{2}\left[-\left(\frac{K_1}{c\,F_1} + 2\alpha_2\right) + \sqrt{\left(\frac{K_1}{c\,F_1} + 2\alpha_2\right)^2 + \frac{4K_1(1-\alpha_2)}{c\,F_1}}\,\right], \tag{7a}$$

$$\alpha_2 = \frac{1}{4}\left[-\alpha_1 + \sqrt{\alpha_1^2 + \frac{8K_2\alpha_1}{c\,F_2}}\,\right]. \tag{7b}$$

In very dilute solutions, as considered in this study, the activity coefficients f_j of the different ions can be approximated by the Debye-Hückel expression ($j = \mathrm{H^+}$, $\mathrm{HSucc^-}$ and $\mathrm{Succ^{2-}}$)

$$\log f_j = -\frac{A\,z_j^2\sqrt{I}}{1 + a_j B\sqrt{I}}, \tag{8}$$

where a_j is the average cation-anion distance (distance of closest approach) and the ionic strength is $I = c(\alpha_1 + 3\alpha_2)$. At 298.15 K the constants are [14] $A = 0.5115\ \mathrm{mol^{-1/2}\,dm^{3/2}}$ and $B = 0.3291 \cdot 10^8\ \mathrm{mol^{-1/2} \cdot dm^{3/2}\ cm^{-1}}$. The activity coefficient of undissociated succinic acid is assumed to be unity.

Molecular conductances Λ are the sum of the ionic contributions:

$$\Lambda = \frac{1000\,L}{c} = \sum_{j=1}^{n} \frac{|z_j|\,c_j\,\lambda_j}{c}, \tag{9}$$

where L is the measured specific conductance and λ_j, c_j and z_j are the individual conductances, concentrations and valences of the ions.

In the case under consideration we have

$$\Lambda = \alpha_1[\lambda(\mathrm{H^+}) + \lambda(\mathrm{HSucc^-})] + 2\alpha_2[\lambda(\mathrm{H^+}) + \lambda(\tfrac{1}{2}\,\mathrm{Succ^{2-}})]. \tag{10}$$

The equation for λ_j exact up to the linear term in the ionic strength I, has the form

$$\lambda_j = \lambda_j^0 - S_j\sqrt{I} + E_j I \ln I + J_{1j} I\,. \tag{11}$$

For unsymmetrical electrolytes, the coefficients S_j, E_j, and J_{1j} are available from the Quint-Viallard theory [15, 16], and with $E_j = 0$ and $J_{1j} = 0$ (11) reduces to the Onsager limiting equation [17]. At given temperature these coefficients depend on the limiting conductances of the corresponding ions, on the viscosity and the dielectric constant of water; the linear term in (11) depends also on the distance of closest approach a_j. The complete expressions for the coefficients are presented in [1, 15, 16].

When expressed in terms of the two step dissociation model, the experimental (Λ, c) data require an adjustment or preselection of six parameters. These are the equilibrium constants K_1 and K_2, the limiting conductances of the anions, $\lambda^0(\mathrm{HSucc^-})$ and $\lambda^0(\tfrac{1}{2}\,\mathrm{Succ^{2-}})$, and the distances of closest approach a_1 and a_2. The values of a_j are equal for the activity coefficient and conductance equations, (8) and (11).

4. Results and Discussion

The equivalent conductances of di-sodium succinate and the molecular conductances of succinic acid are presented in Table 1 and in Figs. 1 and 2. The conductances are plotted as functions of the square root of the ionic strength I, which for a completely dissociated

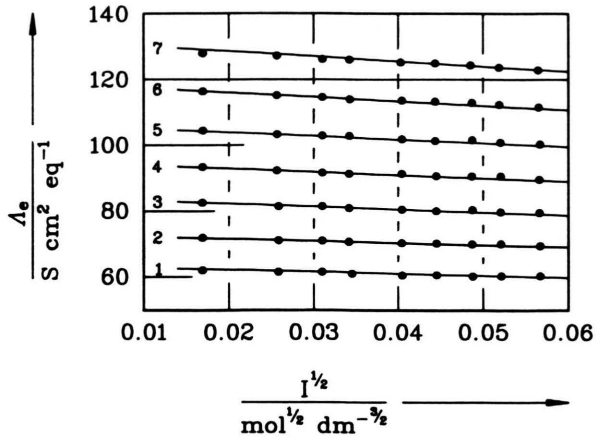

Fig. 1. Equivalent conductances of di-sodium succinate as functions of the square root of ionic strength I.
1: 278.15 K; 2: 283.15 K; 3: 288.15 K; 4: 293.15 K;
5: 298.15 K; 6: 303.15 K; 7: 313.15 K.

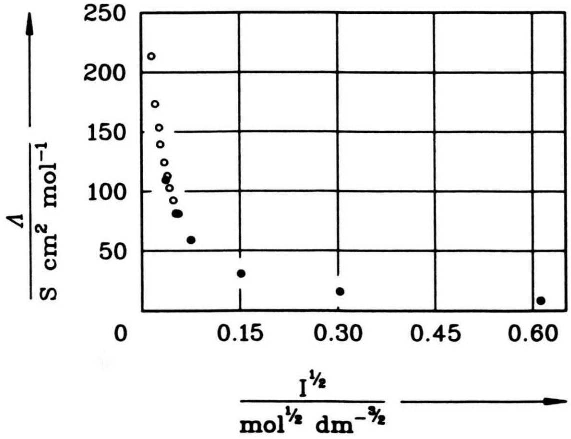

Fig. 2. Molar conductances of succinic acid as a function of the square root of ionic strength at 298.15 K. ●: Jones [5], ○: this work.

1:2 electrolyte is $I = 3c$. As can be seen, di-sodium succinate is a completely dissociated electrolyte and succinic acid is a partially dissociated weak electrolyte. In Fig. 2 the results of Jones [5] are also presented, which cover more concentrated solutions of succinic acid. As many old conductances, they are systematically lower than those determined in modern experiments; the differences are significant especially in dilute solutions. Taking this into account, both sets of data are in reasonable agreement (Figure 2). The units of the equivalent and molar conductances are S cm^2 equiv^{-1} and S cm^2 mol^{-1}; they will be omitted hereafter.

Table 1. Experimental equivalent conductances of di-sodium succinate and molar conductances of succinic acid in the investigated temperature range.

$m \cdot 10^4$ mol kg^{-1}	T/K						
	287.15	283.15	288.15	293.15	298.15	303.15	308.15
$\Lambda_e(\mathrm{Na_2Succ})$							
0.9619	62.18	71.88	82.20	93.02	104.38	116.01	127.96
2.2365	61.61	71.22	81.44	92.17	103.44	115.21	127.43
3.2341	61.28	70.84	81.02	91.67	102.87	114.59	126.69
3.9358	61.03	70.57	80.68	91.30	102.57	114.21	126.31
5.4886	60.57	70.03	80.10	90.64	101.75	113.26	125.21
6.6359	60.35	69.75	79.74	90.24	101.26	112.67	124.69
7.9421	59.98	69.36	79.29	89.72	100.81	112.22	124.08
9.0583	59.46	68.76	78.60	88.94	99.80	111.07	122.84
10.7063	59.27	68.52	78.33	88.61	99.41	110.64	122.34
$\Lambda\,(\mathrm{H_2Succ})$							
0.9171	144.90	162.25	179.68	196.73	213.60	230.00	245.93
1.7091	116.52	130.78	145.07	159.21	173.50	187.10	200.50
2.3665	103.29	116.02	129.71	141.40	153.84	166.01	177.84
3.0613	93.54	105.09	116.68	128.16	139.52	150.61	161.47
4.0330	83.43	93.80	104.16	114.47	124.64	134.55	144.20
5.1799	75.29	84.68	94.11	103.46	112.67	121.69	130.44
6.3613	68.72	77.33	85.96	94.52	103.04	111.29	119.34
7.8857	62.63	70.50	78.37	86.20	93.89	101.51	108.80
10.7880	54.75	61.62	68.54	75.41	82.18	88.81	95.27

The equivalent conductance of completely dissociated electrolytes of the type 1:2, in the Onsager approximation [17] is given by

$$\begin{aligned} \Lambda_e &= \Lambda^0 - S\sqrt{I}\,, \\ \Lambda^0 &= \lambda^0(\mathrm{Na^+}) + \lambda^0(\tfrac{1}{2}\,\mathrm{Succ^{2-}}), \\ S &= \alpha\Lambda^0 + \beta, \end{aligned} \tag{12}$$

where

$$\begin{aligned} \alpha &= \frac{4.8047 \cdot 10^6}{(DT)^{3/2}}\,\frac{q}{1+\sqrt{q}}\,, \\ \beta &= \frac{164.954}{\eta(DT)^{1/2}}\,, \\ q &= \frac{2}{3}\left\{\frac{\lambda^0(\mathrm{Na^+}) + \lambda^0(\frac{1}{2}\,\mathrm{Succ^{2-}})}{2\lambda^0(\mathrm{Na^+}) + \lambda^0(\frac{1}{2}\,\mathrm{Succ^{2-}})}\right\} \end{aligned} \tag{13}$$

and D and η denote the dielectric constant and viscosity of water.

At a given temperature T, the measured equivalent conductances Λ_e can be approximated by a straight line (Fig. 1) and the slopes of these lines are very close to the Onsager slopes (Table 2). The Onsager slopes are based on $\lambda^0(\mathrm{Na^+})$, $D(T)$, $\eta(T)$ values from [14, 17]. The intercepts of the straight lines serve as the limiting equivalent conductances of di-sodium succinate.

Table 2. Limiting equivalent conductances of succinate and sodium ions, the Walden products and the ratios of the observed to the Onsager slopes.

T/K	$\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-})$	$\lambda^0(\mathrm{Na}^+)$	$\lambda^0\eta$	$S_{\mathrm{obs}}/S_{\mathrm{Onsager}}$
278.15	33.01	30.30	0.501	1.007
283.15	38.31	34.88	0.502	0.999
288.15	43.98	39.72	0.503	0.999
293.15	49.92	44.81	0.504	0.996
298.15	56.20	50.15	0.503	1.000
303.15	62.89	56.72	0.503	1.018
308.15	69.55	61.53	0.503	1.001

From the Kohlrausch law the limiting conductances were calculated (Table 2), and at 298.15 K we have $\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) = 56.2$. This result is in a fair agreement with the Davies value [2], $\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) = 57.4$; the Jeffery and Vogel limiting conductance is much higher, $\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) = 60.9$ [6]. The Walden product is practically independent of temperature, $\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) \cdot \eta(T) = 0.503 \pm 0.001$.

The limiting conductance of the hydrosuccinate ion, $\lambda^0(\mathrm{HSucc}^-)$, cannot be determined directly from conductance measurements; its value must be properly assigned. From an analysis of the ammonium hydrogen succinate and sodium hydrogen succinate conductances, the values of $\lambda^0(\mathrm{HSucc}^-) = 31.9$ and $\lambda^0(\mathrm{HSucc}^-) = 35.8$ were adopted by Davies [2] and Topp and Davies [7]. The ratio of limiting conductances varies from system to system and is usually found between 0.5 and 1.2 [18]. Recently, Pethybridge [19] suggested to fix the ratio at 0.80. Evidently, with increasing succinic acid concentration, the contribution due to the second dissociation step is less and less important and the problem can be reduced to the case of a weak, monobasic acid. In the MacInnes and Shedlovsky procedure [20], the degree of dissociation of the primary step is given by

$$\alpha_1 = \Lambda/\Lambda^*, \tag{14}$$

where Λ^* is the conductance of the hypothetically completely dissociated acid (to the primary dissociation step only) at the same concentration c as for the measured conductance Λ. Values of Λ^* can be calculated from

$$\Lambda^* = \Lambda(\mathrm{HCl}) - \Lambda(\mathrm{NaCl}) + \Lambda_{\mathrm{e}}(\mathrm{Na_2Succ})\,. \tag{15}$$

At 298.15 K, the following expressions for the conductances of HCl and NaCl were used [21]:

$$\Lambda(\mathrm{HCl}) = 426.04 - 156.70\sqrt{c} + 165.5\,c(1 - 0.2274\sqrt{c}), \tag{16a}$$

$$\Lambda(\mathrm{NaCl}) = 126.42 - 88.53\sqrt{c} + 89.5\,c(1 - 0.2274\sqrt{c}) \tag{16b}$$

and from this investigation

$$\Lambda_{\mathrm{e}}(\mathrm{Na_2Succ}) = 106.35 - 195.65\sqrt{c}\,. \tag{17}$$

From (14), (15), (16), and (17), for the highest concentration in this work, $c = 1.0675 \cdot 10^{-3}\,\mathrm{mol\,dm^{-3}}$, we have $\Lambda^*(\mathrm{Na_2Succ}) = 397.40$ and $\alpha_1 = 0.2068$, and from

$$K_{11} = \frac{c\,\alpha_1^2 F_1}{1 - \alpha_1} \tag{18}$$

the estimated value of the equilibrium constant is $K_{11} = 5.6 \cdot 10^{-5}$. This value may be compared with dissociation constants of succinic acid reported in the literature, which vary from $K_1 = 6.03$ to $7.24 \cdot 10^{-5}$ and from $K_2 = 1.90$ to $2.47 \cdot 10^{-6}$ [8, 9]. The Pinching and Bates constants are $K_1 = 6.21 \cdot 10^{-5}$ and $K_2 = 2.31 \cdot 10^{-6}$ [10, 11]. The activity coefficient quotient F_1 was determined from the individual activity coefficients (8). Using $\alpha_1 = 0.2068$, $\Lambda = 82.18$, and $\lambda^0(\mathrm{H}^+) = 349.85$, the Quint-Viallard equations (11) for $\Lambda^* = \Lambda_1 = \lambda(\mathrm{H}^+) + \lambda(\mathrm{HSucc}^-)$ were solved simultaneously with (14) to yield the limiting conductance of $\lambda^0(\mathrm{HSucc}^-) = 49.75$, $[\lambda^0(\mathrm{HSucc}^-)/\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) = 0.89]$. Evidently, this result is much higher than that adopted by Davies [2] and Topp and Davies [7].

It follows from (10) that the measured conductance of succinic acid Λ as a function of concentration c is

$$\Lambda = \alpha_1 \Lambda_1 + 2\alpha_2 \Lambda_2\,, \tag{19a}$$

$$\Lambda_1 = \alpha_1[\lambda(\mathrm{H}^+) + \lambda(\mathrm{HSucc}^-)]\,, \tag{19b}$$

$$\Lambda_2 = 2\alpha_2[\lambda(\mathrm{H}^+) + \lambda(\tfrac{1}{2}\,\mathrm{Succ}^{2-})]\,, \tag{19c}$$

where α_1 and α_2 are given by (7) and the individual conductances are calculated from the Quint-Viallard equations (11). The value of $a(\mathrm{Succ}^{2-}) = 8.0$ Å was selected, but the final results are not sensitive to the choice of the distances of closest approach. Considering (19), it is assumed that the contributions coming from the binary electrolyte constituents are additive. Formally, in the limit at infinite dilution (α_1 tends to zero and α_2 approaches unity) we have $\Lambda = 2\Lambda_2$, and this is the molecular conductance of a fully dissociated electrolyte of the type 1:2.

Thus, (10) and (11) tend to the Onsager approximation (12).

Since Λ_1, Λ_2, α_1, α_2, F_1, F_2, and $I = c(\alpha_1 + 3\alpha_2)$ depend on the equilibrium constants K_1 and K_2, it follows from (19) that, since $\Lambda(c; K_1, K_2)$, for a known set of (Λ, c) values the dissociation constants can be approximated with the help of an appropriate iteration procedure. If the equilibrium constants K_1 and K_2 are known at a given concentration c, then α_1, α_2, F_1 and F_2 can consecutively be evaluated from (7), starting with initial values $\alpha_2=0$ and α_1 calculated from (18) with $F_1=1$. The calculations are continued until repetition does not change the α_j and f_j values. Few runs were sufficient in most cases to complete the calculations. α_1 and α_2 determined in this way permit the evaluation of the ionic strength I and finally of Λ_1 and Λ_2. The search for the best agreement between the measured and calculated (19) values of Λ will therefore produce a set of the selfconsistent equilibrium constants. The limiting conductance at 298.15 K can be used for the evaluation of $\lambda^0(\mathrm{HSucc}^-)$ at other temperatures from the Walden rule:

$$\lambda^0(\mathrm{HSucc}^-, T) = \lambda^0(\mathrm{HSucc}^-, 298.15\ \mathrm{K}) \frac{\eta(298.15\ \mathrm{K})}{\eta(T)}. \tag{20}$$

The temperature dependence of $\lambda^0(\mathrm{H}^+)$ and the constants A and B (8) are known [14, 17] and the distances of closest approach, a_j, can be assumed to be independent of temperature.

At 298.15 K, the determined "best" set of parameters is: $\lambda^0(\mathrm{HSucc}^-) = 47.0$, $\lambda^0(\mathrm{HSucc}^-)/\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) = 0.84$, $K_1=5.69 \cdot 10^{-5}$ and $K_2=5 \cdot 10^{-8}$, and the average standard deviation is $\sigma=0.23\ \mathrm{S\ cm^2\ mol^{-1}}$. Since the equilibrium constants, especially K_2, are significantly lower than those reported in the literature, three additional data sets were considered. If the Davies value, $\lambda^0(\mathrm{HSucc}^-) = 31.9$, $\lambda^0(\mathrm{HSucc}^-)/\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) = 0.57$, is selected, then we have from (19): $K_1=6.17 \cdot 10^{-5}$ and $K_2=0.69 \cdot 10^{-6}$ with $\sigma=0.28\ \mathrm{S\ cm^2\ mol^{-1}}$. The choice of the Topp and Davies value, $\lambda^0(\mathrm{HSucc}^-)=35.8$, $\lambda^0(\mathrm{HSucc}^-)/\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) = 0.64$, gives $K_1=6.05 \cdot 10^{-5}$ and $K_2=0.49 \cdot 10^{-6}$ with $\sigma=0.26\ \mathrm{S\ cm^2\ mol^{-1}}$. Finally, if the Pinching and Bates dissociation constants are introduced we have: $\lambda^0(\mathrm{HSucc}^-)=22.9$, $\lambda^0(\mathrm{HSucc}^-)/\lambda^0(\frac{1}{2}\,\mathrm{Succ}^{2-}) = 0.41$, with $\sigma=1.38\ \mathrm{S\ cm^2\ mol^{-1}}$. The measured and calculated values of Λ for these four sets of parameters are presented in Table 3. As can be seen, in the framework of the Quint and Viallard theory there is no satisfactory description of the observed conductances with regard to a proper choice of $\lambda^0(\mathrm{HSucc}^-)$. Preselection of the dissociation constants gives results which are clearly inconsistent with the accuracy of our experiments. The "best" set of parameters is practically equivalent to the case of a monobasic weak acid with a very small correction due to the second dissociation step. This disagrees with the accepted values of K_1 and K_2 for succinic acid [8, 9]. Considering K_2, the selection of the Davies limiting conductance is somewhat better than that chosen by Topp and Davies. However, there is no clear distinction between these two cases, because the calculations are rather insensitive to changes in $\lambda^0(\mathrm{HSucc}^-)$. The same picture is observed at other temperatures. At this stage of investigation, without knowledge of transference numbers, it is difficult to assign properly the limiting conductance of the ion HSucc^-; only accurate $\lambda^0(\mathrm{HSucc}^{2-})$ values as a function of temperature are available.

Table 3. Experimental and calculated (19) molar conductances of succinic acid at 298.15 K.

$c \cdot 10^4$/ mol dm^3	Λ_{exp}	Λ_{calc} [a]	Λ_{calc} [b]	Λ_{calc} [c]	Λ_{calc} [d]
0.9144	213.60	213.83	213.97	213.88	216.76
1.7040	173.50	173.26	173.21	173.33	173.64
2.3596	153.84	153.65	153.59	156.63	153.36
3.0523	139.52	139.14	139.09	139.13	138.52
4.0212	124.64	124.66	124.63	124.67	123.82
5.1647	112.67	112.50	112.48	112.53	111.55
6.3427	103.04	103.21	103.21	103.26	102.22
7.8627	93.89	94.17	94.19	94.23	93.17
10.7563	82.18	82.17	82.20	82.23	81.19

[a] $\lambda^0(\mathrm{HSucc}^-)=47.0$, $K_1=5.69 \cdot 10^{-5}$ and $K_2=5 \cdot 10^{-8}$.
[b] $\lambda^0(\mathrm{HSucc}^-)=31.9$ [2], $K_1=6.17 \cdot 10^{-5}$ and $K_2=0.69 \cdot 10^{-6}$.
[c] $\lambda^0(\mathrm{HSucc}^-)=35.8$ [7], $K_1=6.05 \cdot 10^{-5}$ and $K_2=0.49 \cdot 10^{-6}$.
[d] $\lambda^0(\mathrm{HSucc}^-)=22.9$, $K_1=6.21 \cdot 10^{-5}$ and $K_2=2.31 \cdot 10^{-6}$ [10, 11].

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