

Diffusion Coefficients for the System Potassium Chromate–Water at 25 °C

Nicola Iadicicco, Luigi Paduano, and Vincenzo Vitagliano*

Dipartimento di Chimica, Università di Napoli, Federico II, Via Mezzocannone 4, 80134 Napoli, Italy

Accurate diffusion coefficients, densities, and viscosities were measured for the aqueous solutions of potassium chromate at 25 °C over a wide range of concentration. Using conductivity, transference numbers, and activity coefficient data from the literature, the ionic phenomenological mobilities were computed. The results are briefly discussed.

Introduction

Experimental diffusion coefficients of various aqueous electrolyte solutions are necessary in order to understand and model a variety of chemical, geochemical (Anderson and Graf, 1978; Berner 1980), and industrial (Tuwiner, 1962) processes.

Transport properties of 2:1 electrolyte solutions are lacking in the literature, in spite of the importance of most of these compounds.

Among them, not much information can be found about chromate salts. These compounds are used in the tanning industry and are the source of the major pollution of waste waters from these industries. A knowledge of the transport properties in binary and higher number components of chromate salts is important.

We chose potassium chromate–H₂O as a model system to start a diffusion study in this field. No diffusion data measured with interferometric techniques were found in the literature for K₂CrO₄ aqueous solutions. Lobo (1989) reports some diffusion data on this system; the only one comparable with ours is about 2.5% higher (see Figure 2). A set of K₂CrO₄ diffusion data measured at 25 °C with the Taylor dispersion technique was published by Vacek and Rod (1986), but we could not obtain their original paper and compare their data with ours.

K₂CrO₄ is slightly hydrolyzed in aqueous solutions; however, from pH measurements we found that the relative concentration of OH[−] ions is always lower than $\sim 5 \times 10^{-4}$ mol dm^{−3}, in the concentration range of our diffusion measurements; so we neglected its possible contribution to the diffusion process.

Only very old data were found in the literature about properties connected with diffusion for K₂CrO₄ solutions (Lobo, 1984). Jones and Jacobson published conductance measurements at 25 °C in 1908; their data turned out to be accurate and in very good agreement with our diffusion data, as will be shown later.

We fitted their molar conductivity data to the following equation:

$$\Lambda/S \text{ cm}^2 \text{ mol}^{-1} = 290.2 - 666.3c^{1/2} + 1502.4c - 1335.8c^{3/2} \pm 0.8 \quad (1)$$

where $c/\text{mol dm}^{-3}$ is the molar concentration and Λ is the molar conductivity. Equation 1 fits the experimental data up to $c \cong 0.25 \text{ mol dm}^{-3}$. From eq 1 the limiting molar conductivity of K₂CrO₄ was assumed to be $\Lambda^\circ = 290.2 \text{ S cm}^2 \text{ mol}^{-1}$ and from the limiting conductivity of potassium

ion, as given by Harned and Owen (1958) [Table 6-8-2], $\lambda_+^\circ = 73.52 \text{ S cm}^2 \text{ mol}^{-1}$, we obtained the limiting molar conductivity of the CrO₄^{2−} ion, $\lambda_-^\circ = 143.2 \text{ S cm}^2 \text{ mol}^{-1}$, and the limiting transport numbers of K⁺ and CrO₄^{2−}: $t_+^\circ = 0.507$ and $t_-^\circ = 0.493$.

Transport number data on K₂CrO₄ aqueous solutions are listed by Lobo (1984). We choose the two Hittorf data sets published by Steel (1902) because the straight line passing through them as a function of the square root of concentration fits the limiting value obtained from conductivity:

$$t_- = 0.4927 - 0.090c^{1/2} \pm 0.0005 \quad (c \leq 1 \text{ mol dm}^{-3}) \quad (2)$$

Activity coefficients γ_\pm of K₂CrO₄ aqueous solutions are given by Harned and Owen (1958) [Table 13-1-2A]. We fitted these data to the following extended Debye–Hückel expression:

$$\ln \gamma_\pm = -4.0547m^{1/2}/(1 + 2.1m^{1/2}) - 0.1275m + 0.04431m^2 - \ln(1 + 0.05405m) \quad (3)$$

Equation 3 reproduces the activity coefficients within the third decimal figure, for $m \leq 2 \text{ mol kg}^{-1}$.

Experimental Section

Materials. Carlo Erba K₂CrO₄ (assay >99.5%) was used without further purification for all experiments. Two stock solutions of K₂CrO₄ + H₂O were prepared, filtered, and used for all experiments. In all calculations the molecular weights of K₂CrO₄ and water were assumed to be 194.20 and 18.016, respectively.

The compositions of the stock solutions were determined by volumetric titration with the Mohr salt of known masses of stock solutions; the standard deviation on their composition was found to be about 0.2%. The data on these solutions are given in Table 1.

Density Measurements. All solutions were prepared by weighing the stock solution samples and the doubly-distilled water used for dilution. To convert mass concentrations to volume concentrations, densities were measured for all solutions with a Anton Parr DMA 602. An accuracy of $\pm 3.0 \times 10^{-5} \text{ g cm}^{-3}$ was obtained. The temperature of the density meter was regulated at $(25.00 \pm 0.01) \text{ }^\circ\text{C}$ and calibrated with air and water where the density of air-saturated water was assumed to be $0.997043 \text{ g cm}^{-3}$. Table 1 collects all the density results.

Viscosity. Viscosity measurements were performed at $(25 \pm 0.02) \text{ }^\circ\text{C}$ with an Ubbelohde viscometer having a

* FAX: (3981) 5527771. E-mail: VITA@CHEMNA.DICHI.UNINA.IT.

Table 1. Densities of Potassium Chromate Aqueous Solutions at 25°C

$m/\text{mol kg}^{-1}$	$\rho/\text{g cm}^{-3}$	$m/\text{mol kg}^{-1}$	$\rho/\text{g cm}^{-3}$	$m/\text{mol kg}^{-1}$	$\rho/\text{g cm}^{-3}$
0.000 00	0.997 043	0.4055	1.056 517	1.5571	1.203 057
7.00×10^{-6}	0.997 047	0.4632	1.064 602	1.6656 ^a	1.215 470
0.003 50	0.997 576	0.4914	1.068 524	1.6945	1.218 833
0.005 23	0.997 834	0.5137	1.071 601	1.7513	1.225 185
0.017 57	0.999 761	0.5434	1.075 677	1.8836	1.239 756
0.019 83	1.000 072	0.6450	1.089 466	1.9403	1.245 937
0.039 31	1.003 045	0.7076	1.097 833	2.0723	1.260 082
0.040 51	1.003 258	0.7437	1.102 621	2.1288	1.266 022
0.057 09	1.005 745	0.7790	1.107 277	2.2558	1.279 213
0.058 02	1.005 886	0.8639	1.118 383	2.3215	1.285 914
0.059 18	1.006 062	0.9261	1.126 387	2.4436	1.298 263
0.063 44	1.006 706	1.1699	1.156 964	2.5092	1.304 737
0.097 47	1.011 835	1.2048	1.161 244	2.6308	1.316 693
0.126 69	1.016 205	1.3974	1.184 415	2.6963	1.323 011
0.160 05	1.021 166	1.4147	1.186 445	2.8153	1.334 287
0.2001	1.027 051	1.4328	1.188 589	2.8853	1.340 909
0.2324	1.031 777	1.4410	1.189 555	2.9155	1.343 712
0.2544	1.034 968	1.4622	1.191 963	2.9714	1.348 817
0.2772	1.038 265	1.4910	1.195 408	3.1263	1.363 045
0.2844	1.039 307	1.4987	1.196 305	3.1959 ^a	1.369 202
0.3362	1.046 724	1.5097	1.197 582		
0.3833	1.053 418	1.5290	1.199 823		

y	A_0	A_1	$A_2 \times 10^2$	$A_3 \times 10^3$	$A_4 \times 10^3$	$A_5 \times 10^4$	$A_6 \times 10^5$	$\epsilon \times 10^5$
$\rho^b/\text{g cm}^{-3}$	0.997 043	0.153 501	-1.858 54	5.373 85	-1.9306	3.925 09	-3.137 66	2.6
$c^b/\text{mol dm}^{-3}$		0.997 326	-4.198 62	-6.527 38	2.8055	-5.904 74	5.525 88	3.7

^a Stock solutions. ^b Densities and concentrations were fitted by the following equation:

$$y = \sum_{i=0}^6 A_i m^i \pm \epsilon \quad (6)$$

Table 2. Viscosities of K₂CrO₄ Aqueous Solutions at 25 °C

$c/\text{mol dm}^{-3}$	t/t^0	$\rho/\text{g cm}^{-3}$	η/η^0
2.700	1.362	1.369 23	1.870
2.542	1.319	1.348 95	1.785
2.401	1.283	1.330 72	1.712
2.275	1.253	1.314 32	1.652
2.058	1.209	1.285 98	1.559
1.879	1.176	1.262 33	1.489
1.729	1.150	1.242 27	1.433
1.601	1.130	1.225 07	1.388
1.490	1.114	1.210 13	1.352
1.394	1.101	1.197 04	1.322
1.271	1.088	1.180 19	1.288
1.161	1.074	1.164 98	1.255
0.996	1.058	1.142 11	1.212
0.934	1.051	1.133 38	1.195
0.821	1.041	1.117 48	1.167
0.775	1.039	1.110 94	1.158
0.634	1.029	1.090 85	1.126
0.526	1.023	1.075 23	1.103
0.409	1.017	1.058 24	1.079
0.283	1.012	1.039 74	1.055
0.160	1.006	1.021 38	1.031
0.092	1.005	1.011 11	1.019
0.000	1.000	0.997 04	1.000

$t^0 = 204.13$ s

$\eta/\eta^0 = 1 + 0.1914c - 0.00205c^2 + 0.0250c^3 - 0.00238c^4 \pm 0.0013$

water running time $t^0 = 204.13$ s. The experimental results are given in Table 2.

Our density and viscosity data are in good agreement with previously published values by Jones and Colvin (1940). There is a slight systematic difference between our data and the previous ones, probably due to the different way of preparing the solutions (see Figure 1).

Diffusion Measurements. The experiments were performed with a Gouy diffusometer (Gosting, 1950; Tyrrell and Harris, 1984), which has been automated to scan Gouy fringe patterns and record fringe positions during an

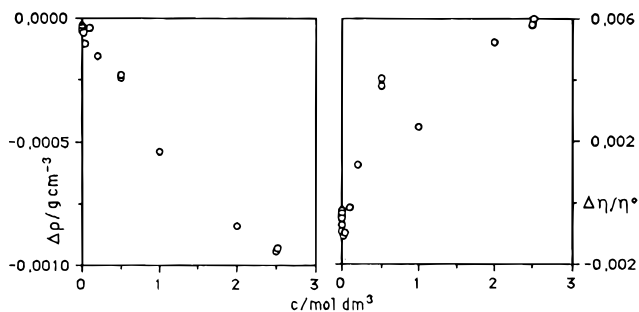


Figure 1. Comparison between our density and viscosity data and those published by Jones and Colvin (1940): (Δy) difference between our data and the previous data.

experiment. A Model "II fx" MacIntosh computer was used to control the scanning apparatus and to calculate fringe positions from fringe intensity profiles. The Gouy diffusometer is a two-lens apparatus in which parallel light is focused through the diffusion cell. The light source is a Unifas PHA SE 0.8-mW neon-helium laser operating at $\lambda = 632.8$ nm. In this parallel light arrangement, the b distance of the apparatus is the distance from the second principal plane of the lens on the detector side of the apparatus to the plane in which the detector scans the patterns; $b = 193.39$ cm. The temperature was regulated at (25.00 ± 0.01) °C. Some further details on our diffusometer are given by Paduano et al. (1992).

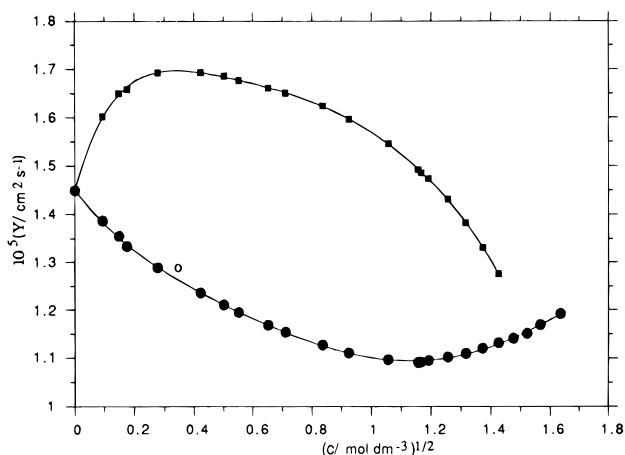
All diffusion results are given in Table 3. Because of the possible impurities in K_2CrO_4 , the uncertainty in the values of D should be considered to be $\pm 0.3\%$ or less as a possible systematic error. Figure 2 is a graph of the measured diffusion coefficients drawn as a function of the square root of concentration. The mobility, eq 10, is also shown in Figure 2.

The value of the diffusion coefficient at infinitesimal concentration was calculated from the Nernst-Hartley equation (Harned and Owen, 1958) [eq 6-10-7], by using

Table 3. Diffusion Data on the System Potassium Chromate–Water at 25 °C

$c/\text{mol dm}^{-3}$	$\Delta c/\text{mol dm}^{-3}$	J_m	$10^5 D/\text{cm}^2 \text{ s}^{-1}$
0.0000			1.4494
0.00876	0.0175	28.12	1.387
0.02193	0.0368	58.70	1.355
0.03103	0.0516	81.55	1.334
0.07728	0.0391	59.89	1.289
0.1782	0.0393	60.13	1.236
0.2513	0.0437	66.01	1.211
0.3052	0.0502	75.54	1.195
0.4248	0.0554	81.84	1.169
0.5041	0.0493	72.09	1.154
0.6998	0.0335	48.39	1.128
0.8558	0.0567	80.21	1.111
1.1184	0.0308	42.15	1.092
1.3371	0.0407	55.11	1.092
1.3608	0.0428	56.98	1.092
1.4182	0.0402	54.04	1.095
1.5775	0.0471	62.96	1.102
1.7329	0.0461	60.34	1.110
1.8844	0.0449	57.87	1.120
2.0323	0.0510	65.45	1.131
2.1764	0.0497	62.97	1.141
2.3170	0.0486	60.38	1.152
2.4541	0.0509	63.04	1.169
2.6759	0.0486	59.12	1.192

^a c , average concentration. Δc , concentration difference between bottom and top solutions of each run. J_m , number of Gouy fringes, in terms of refractive index difference, between bottom and top solutions at the He–Ne laser red light: $J_m = 3.951 \times 10^6 \Delta n$. D , diffusion coefficient.

**Figure 2.** Diffusion coefficients, D (●), and mobilities, \mathcal{M} (■), of K_2CrO_4 aqueous solutions at 25 °C; (○) from Lobo (1989, p 939).

the given limiting ionic conductances:

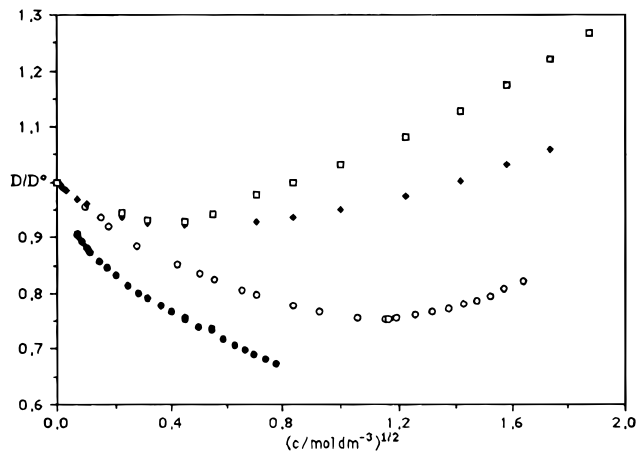
$$D^0/\text{cm}^2 \text{ s}^{-1} = 8.936 \times 10^{-10} T \frac{\nu_+ + \nu_- (\lambda_+^0 \lambda_-^0)}{\nu_+ |z_+|} \left(\frac{\Lambda^0}{\Lambda^0} \right) = 1.4494 \times 10^{-5} \quad (4)$$

where ν_i is the stoichiometric coefficient of ion i in the salt and z_i is its charge number. This value was found in perfect agreement with our experimentally measured value, confirming the goodness of Jones and Jacobson (1908) conductance measurements; in fact the fitting of the first five diffusion runs of Table 3 as a function of $c^{1/2}/(\text{mol cm}^{-3})^{1/2}$ gives the following expression:

$$10^5 D/\text{cm}^2 \text{ s}^{-1} = 1.4496 - 0.733c^{1/2} + 0.539c \quad (5)$$

Discussion

Figure 3 is a comparison of relative diffusion coefficients for some 1:1 and 2:1 potassium salts. It shows that, at

**Figure 3.** Relative diffusion coefficients, D/D^0 , for various potassium salts: (□) KI, $D^0 = 2.001 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ (Dunlop and Stokes, 1951); (◆) KCl, $D^0 = 1.993 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ (Harned and Nuttal, 1949; Gosting, 1950); (○) K_2CrO_4 , $D^0 = 1.450 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$; (●) K_2SO_4 , $D^0 = 1.530 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ (Miller and Rard).

finite concentrations, diffusion coefficients increase with increasing molecular weight: $D_{\text{KI}} > D_{\text{KCl}}$. In particular, $D_{\text{K}_2\text{CrO}_4} > D_{\text{K}_2\text{SO}_4}$. The reason for this behavior may be due to the smaller hydration shell of the anion; as its dimension increases, so can the anion diffuse in a more destructured solvent environment. However, the limiting diffusion coefficient of K_2SO_4 is higher than that of K_2CrO_4 .

A whole description of the phenomenology of transport in a $(\nu_+ - \nu_-)$ -electrolyte solution, in terms of irreversible thermodynamics, was published by Miller (1966). The author stressed the necessity of defining three transport coefficients to fully describe this process.

The phenomenological equations can be written as

$$J_1 = \dot{\iota}_{11} X_1 + \dot{\iota}_{12} X_2 \quad (6a)$$

$$J_2 = \dot{\iota}_{21} X_1 + \dot{\iota}_{22} X_2 \quad (6b)$$

where indices 1 and 2 refer to cations and anions, respectively, and the driving forces are

$$X_i = - \left[\frac{\partial \mu_i}{\partial x} + z_i \mathcal{F} \frac{\partial \phi}{\partial x} \right] \quad (7)$$

where μ_i is the chemical potential of ion i , \mathcal{F} is the faraday constant, and ϕ the electrical potential.

If the flows J_i and the phenomenological coefficients $\dot{\iota}_{ij}$ are expressed in terms of the solvent-fixed reference frame (SF), the entropy production becomes

$$\sigma = J_1 X_1 + J_2 X_2 \quad (8)$$

and the Onsager reciprocal relations hold:

$$\dot{\iota}_{12} = \dot{\iota}_{21} \quad (9)$$

The experimental diffusion coefficients, D , are measured in a volume-fixed reference frame. The mobility is related to either the volume-fixed D or the solvent-fixed D_{SF} by the equation (Miller, 1966)

$$\mathcal{M} = D/(1 + d \ln \gamma_{\pm}/d \ln m) = D_{\text{SF}}/(1 + d \ln y_{\pm}/d \ln c) \quad (10)$$

where γ_{\pm} and y_{\pm} are the activity coefficients in the molal and molar concentration scales, respectively.

Table 4. Thermodynamic Factors, Mobilities, and Ionic Mobilities of Aqueous Solutions of Potassium Chromate at 25 °C^a

<i>m</i> /mol kg ⁻¹	1 + (d ln γ _± /d ln <i>m</i>)	10 ⁵ <i>M</i> /cm ² s ⁻¹	cm ² s ⁻¹ J ⁻¹ × 10 ¹²			<i>m</i> /mol kg ⁻¹	1 + (d ln γ _± /d ln <i>m</i>)m ² s ⁻¹	10 ⁵ <i>M</i> /cm ² s ⁻¹
			<i>λ</i> ₁ /2 <i>c</i>	<i>λ</i> ₂ /2 <i>c</i>	<i>λ</i> ₂₂ /2 <i>c</i>			
0.0000	1.0000	1.4494	7.91	0.00	1.93	1.1874	0.7089	1.547
0.0088	0.8657	1.602	7.73	0.55	1.84	1.4384	0.7314	1.492
0.0220	0.8212	1.650	7.64	0.74	1.80	1.4660	0.7345	1.487
0.0312	0.8040	1.659	7.58	0.81	1.77	1.5334	0.7427	1.474
0.0778	0.7616	1.692	7.48	0.99	1.71	1.7229	0.7696	1.432
0.1801	0.7297	1.694	7.37	1.11	1.64	1.9120	0.8026	1.382
0.2548	0.7185	1.686	7.24	1.17	1.59			
0.3103	0.7126	1.677	7.10	1.23	1.55			
0.4344	0.7033	1.662	6.99	1.27	1.50			
0.5174	0.6992	1.651	6.88	1.31	1.46			
0.7256	0.6944	1.624	6.61	1.38	1.39			
0.8950	0.6957	1.597	6.39	1.42	1.33			

^a *m*, molality of each run. [1 + (d ln γ_±/d ln *m*)], thermodynamic factor. *M*, mobility = *D*[1 + (d ln γ_±/d ln *m*)], eq 10. The values printed in italics correspond to extrapolated values, those at higher concentrations are only indicative.

The explicit expression for the Onsager *λ*_{*ij*} coefficients, as given by Miller (1966), is

$$\frac{\lambda_{ij}}{2c} = \frac{t_i t_j \Lambda / 2}{10^3 \mathcal{F}^2 z_i z_j} + \frac{v_i v_j}{10^3 RT (v_i + v_j) v_1 z_1} \mathcal{M} \quad (11)$$

where *c* is the electrolyte concentration, *RT* has the usual meaning, and the *λ*_{*ij*}/*c* dimensions are mol cm² s⁻¹ J⁻¹.

In terms of numerical values for the K₂CrO₄ solutions

$$\frac{\lambda_{11}}{2c} = 1.0740 \times 10^{-13} t_+^2 \Lambda / 2 + 2.689 \times 10^{-7} \mathcal{M} \quad (12a)$$

$$\frac{\lambda_{12}}{2c} = -0.5370 \times 10^{-13} t_+ t_- \Lambda / 2 + 2.689 \times 10^{-7} \mathcal{M} \quad (12b)$$

$$\frac{\lambda_{22}}{2c} = 0.2685 \times 10^{-13} t_-^2 \Lambda / 2 + 0.6775 \times 10^{-7} \mathcal{M} \quad (12c)$$

where *Λ*/*S* cm² mol⁻¹ and *t*_± are the molar conductivity and transference number taken from eqs 1 and 2.

From eq 11, for *i* = 1 and *j* = 2, since *v*₁*z*₁ = -*v*₂*z*₂, one obtains

$$\mathcal{M} = RT \frac{(v_1 + v_2) t_1 t_2 \Lambda}{2 v_1 z_1 \mathcal{F}^2} + 10^3 RT \frac{(v_1 + v_2) v_1 z_1}{v_1 v_2} \frac{\lambda_{12}}{2c} \quad (13)$$

The first term on the right hand side of eq 13 is the term of the Nernst–Hartley equation for the diffusion coefficient. This term reduces to eq 4 for a 2:1 electrolyte at infinitesimal concentration. As pointed out by Miller (1966) the failure of the Nernst–Hartley equation to describe the diffusion coefficient of electrolytes at finite concentration is due to the lack of the term on the right hand side of eq 13 containing the *λ*₁₂/*c* coefficient.

Table 4 collects all main and ionic mobilities; Figure 4 is a graph of the ionic mobilities drawn as a function of the square root of concentration.

It is interesting to note that, while the mean mobility term, *M*, goes through a maximum, the ionic mobilities *λ*_{*ij*}/*c* all have a monotonic trend; *λ*₁₁/*c* and *λ*₂₂/*c* are regularly decreasing, while *λ*₁₂/*c* increases. This term is eventually expected to go through a maximum at higher concentrations, as found for other electrolytes (Miller, 1966).

At infinitesimal concentration the cross term *λ*₁₂/*c* is zero; as concentration increases, the dragging effect of the ions on those of opposite charge becomes significant so that *λ*₁₂/*c* increases. In 1:1 electrolytes this term is in general reasonably smaller than the main terms *λ*_{*ij*}/*c*; however, in

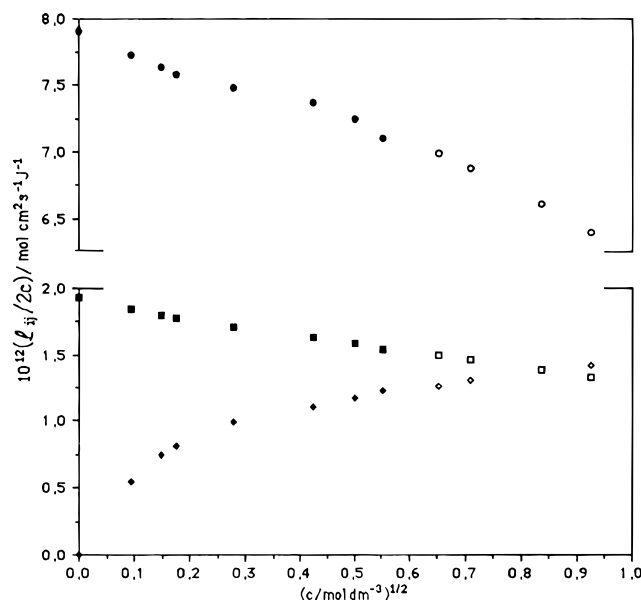


Figure 4. Individual ionic mobilities. Index 1 for K⁺ and index 2 for CrO₄²⁻ from eq 12. Key: (●) *λ*₁₁/2*c*; (◆) *λ*₁₂/2*c*; (■) *λ*₂₂/2*c*. The *λ*_{*ij*}/*c* dimensions are mol cm² s⁻¹ J⁻¹. Open symbols correspond to extrapolated values, only indicative.

K₂CrO₄ solutions the presence of the doubly charged chromate ion makes the dragging effect much stronger so that the *λ*₁₂/*c* values become quite large, and they are expected to become larger than the main term *λ*₂₂/*c* at higher concentration. A similar behavior was found in CuSO₄ solutions where both ions bear a double charge, (Miller et al., 1980).

Not much can be said about viscosity. We measured it mainly with the purpose of collecting most of the lacking phenomenological transport properties of K₂CrO₄ solutions. If mobilities or ionic mobilities are multiplied by the term *η*/*η*⁰, a slightly more regular trend is observed; however, as pointed out by some authors (Miller, 1966), any phenomenological correlation between viscosity and mobilities cannot be made because of the different tensorial nature of these quantities. Viscosity is a second-order tensor, while mobilities are vectors. Any correlation between these quantities should be found in terms of local molecular contributions.

Acknowledgment

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