

The Viscosity of Aqueous KCl Solutions in the Temperature Range 25–200°C and the Pressure Range 0.1–30 MPa

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In this paper, new precise viscosity data are presented for aqueous solutions of KCl; these data cover the temperature range 25–200°C, the pressure range 0.1–30 MPa, and the concentration range 0–4.6 mol KCl/kg H₂O. An empirical correlation which reproduces the data within the experimental precision of $\pm 0.5\%$ is given. The accuracy of the data is estimated to range from ± 0.5 to $\pm 1.5\%$.

KEY WORDS: high pressure; KCl; solutions; viscosity.

1. INTRODUCTION

In two previous publications [1, 2], we have presented data on the viscosity of water and aqueous solutions of NaCl covering the temperature range 25–200°C, the pressure range 0.1–30 MPa and, for the NaCl system, the concentration range 0–6 mol · kg⁻¹. These data were obtained with an oscillating-disk viscometer [1] designed specifically for measurements on corrosive fluids under the conditions of moderately high temperatures and pressures. In this paper, we describe measurements on aqueous KCl solutions over similar temperature and pressure ranges and over the concentration range 0–4.6 mol · kg⁻¹. The behavior of the viscometer in these corrosive environments represents somewhat of an improvement upon an earlier instrument [3], which did not possess the same corrosion resistance. Consequently, the data obtained with the new instrument are to be pre-

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ferred over those obtained with the earlier version [3–5]. The two data sets do, nevertheless, agree within their combined uncertainty limits. The measurements discussed in this paper possess a precision of $\pm 0.1\%$ at 25°C , degrading to $\pm 0.5\%$ at 200°C .

2. EXPERIMENTAL

The design and construction of the oscillating-disk viscometer along with the methods of temperature and pressure measurement have been adequately described in an earlier publication [1]. The characteristics of the oscillating system and the calibration of the instrument with respect to water are identical with those employed in our study of the aqueous NaCl system [2]. As has been our previous practice [2], the calibration of the instrument was checked prior to the start of measurements on a new solution; at no time did these checkpoints deviate by more than $\pm 0.2\%$ from the original calibration. The working equations for the oscillating-disk viscometer, through which the viscosity is determined from measurements of the logarithmic decrement and period of oscillation, have been described previously [1, 3, 6]. More recently, Kestin and Shankland [7] have re-evaluated the relative method of measurement and proposed a more self-consistent working equation. However, in the case of the present measurements, the two working equations yielded indistinguishable results.

Prerequisite to the determination of the viscosity with the oscillating-disk method is a knowledge of the fluid density. Unfortunately, there exist few accurate data on the density of aqueous KCl solutions over the temperature and pressure ranges employed in this study. For the temperature range $25\text{--}150^\circ\text{C}$, we have used the density correlation of Grimes et al. [5]; this correlation is based on an extensive compilation of data assembled by Potter and Brown [8]. For temperatures in excess of 150°C , we have employed the data of Egerov et al. [9].

3. ANALYSIS AND CORRELATION OF RESULTS

The experimental viscosity results are summarized in Tables 1–5; the majority of the data collected in these tables represent the average of two measurements. Experimental data have been corrected to nominal temperatures by means of the relation

$$\eta(t_{\text{nom}}) = \eta(t) + (\partial\eta/\partial t)_p(t_{\text{nom}} - t), \quad (1)$$

where the derivative $(\partial\eta/\partial t)_p$ has been estimated from correlations provided in earlier studies of this system [5, 10, 11]. In the absence of such

Table I. Viscosity of Aqueous KCl (0.99005 mol · kg⁻¹)

<i>P</i> (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)	<i>P</i> (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)
(a) <i>t</i> = 26.0°C		(b) <i>t</i> = 31.8°C	
0.3	869.2	0.3	776.0
5.7	869.5	5.7	776.2
10.9	869.5	10.8	776.8
15.8	869.4	15.7	777.1
21.0	869.6	20.9	778.2
25.9	870.1	26.0	778.8
30.9	870.7	31.0	780.3
0.2	869.8	0.2	776.4
(c) <i>t</i> = 41.8°C		(d) <i>t</i> = 51.3°C	
0.4	648.0	0.6	557.5
5.8	649.0	5.6	558.6
10.8	649.9	10.8	559.9
15.9	651.0	15.8	561.0
20.8	651.8	20.9	562.3
25.9	652.5	25.7	563.1
30.9	653.9	30.8	564.4
0.8	647.4	1.2	557.9
(e) <i>t</i> = 66.0°C		(f) <i>t</i> = 92.0°C	
0.5	452.4	0.6	331.5
5.6	454.0	5.5	333.2
10.5	455.1	10.6	335.2
15.6	456.7	15.6	336.0
21.4	458.3	20.5	336.8
25.5	459.0	25.8	338.6
30.9	460.7	30.6	339.7
1.6	453.1	1.4	332.2
(g) <i>t</i> = 121.2°C		(h) <i>t</i> = 158.8°C	
2.0	251.3	3.1	189.6
7.4	252.8	7.4	190.8
14.2	254.8	13.7	192.1
20.8	256.5	20.2	193.3
25.4	258.2	25.4	194.8
30.3	259.1	30.6	195.8
1.5	250.8	4.0	189.6
(i) <i>t</i> = 199.2°C			
4.3	152.1		
8.7	152.6		
15.4	154.0		
20.6	155.0		
25.8	155.2		
31.2	157.6		

Table II. Viscosity of Aqueous KCl (2.07510 mol · kg⁻¹)

<i>P</i> (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)	<i>P</i> (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)
(a) <i>t</i> = 24.9°C		(b) <i>t</i> = 29.0°C	
0.3	897.1	0.2	831.0
5.4	897.0	5.5	832.4
10.5	897.9	10.6	832.6
15.7	898.9	15.6	833.3
20.8	899.9	20.8	833.9
25.5	899.8	25.9	835.0
30.7	900.7	30.8	835.6
		0.6	831.2
(c) <i>t</i> = 40.1°C		(d) <i>t</i> = 50.0°C	
0.4	687.9	0.3	592.3
5.4	688.8	5.7	594.0
10.5	690.3	10.6	594.7
15.6	691.4	15.5	595.8
20.9	692.6	20.7	597.1
26.0	693.8	26.0	598.6
31.1	695.3	31.3	599.9
0.8	688.6	0.6	592.1
(e) <i>t</i> = 67.3°C		(f) <i>t</i> = 91.5°C	
0.4	470.7	0.9	359.0
5.6	471.7	5.4	360.1
10.5	473.6	10.6	361.5
15.6	474.8	15.1	364.6
21.2	476.5	20.6	365.8
25.8	477.5	25.7	367.7
31.0	479.0	31.0	368.9
1.5	471.1	0.8	360.9
(g) <i>t</i> = 121.2°C		(h) <i>t</i> = 162.1°C	
3.3	276.0	2.3	207.3
8.1	277.6	3.0	206.6
13.5	279.5	8.4	208.4
20.0	281.1	15.4	209.3
25.0	282.9	20.7	210.2
30.7	284.6	25.5	211.4
3.2	276.5	29.7	212.9
		2.9	207.3

Table III. Viscosity of Aqueous KCl (3.10040 mol · kg⁻¹)

<i>P</i> (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)	<i>P</i> (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)
(a) <i>t</i> = 24.4°C		(b) <i>t</i> = 29.3°C	
0.4	929.0	0.3	850.5
5.6	928.8	5.5	851.7
10.7	929.9	10.7	852.9
15.7	931.2	15.4	854.1
20.9	932.3	20.9	855.6
25.8	933.6	26.0	857.4
31.1	934.8	31.0	859.1
0.3	927.9	0.2	851.5
(c) <i>t</i> = 39.8°C		(d) <i>t</i> = 49.0°C	
0.5	718.7	0.3	627.8
5.6	719.5	5.7	628.8
10.4	720.8	10.7	630.3
15.5	722.2	15.6	631.7
20.7	723.5	21.0	633.3
26.0	724.7	25.8	634.5
31.2	726.8	31.1	636.6
0.7	719.0	1.1	627.4
(e) <i>t</i> = 65.5°C		(f) <i>t</i> = 90.9°C	
0.6	508.4	1.0	386.7
5.5	510.1	5.4	388.4
10.7	511.4	10.5	390.5
15.4	512.9	15.5	391.3
20.8	514.3	21.0	393.4
25.6	515.3	26.0	394.9
31.6	516.9	31.1	396.4
1.2	508.5		
(g) <i>t</i> = 121.2°C		(h) <i>t</i> = 162.4°C	
3.2	297.4	3.6	225.9
8.1	299.0	7.2	226.1
14.1	300.9	13.7	228.3
20.5	303.3	19.9	228.3
26.2	304.3	26.2	230.1
31.0	305.7	29.6	231.5
3.6	297.9		
(i) <i>t</i> = 202.2°C			
3.4	184.2		
9.0	185.6		
14.4	185.4		
20.9	188.4		
26.0	188.4		
31.0	190.1		
4.0	184.7		

Table IV. Viscosity of Aqueous KCl ($3.96850 \text{ mol} \cdot \text{kg}^{-1}$)

P (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)	P (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)
(a) $t = 24.9^\circ\text{C}$		(b) $t = 29.3^\circ\text{C}$	
0.4	940.1	0.2	872.6
5.2	941.5	5.2	873.7
10.5	943.3	10.3	875.2
15.4	944.4	15.2	877.2
20.6	946.5	20.3	879.0
25.7	947.1	26.2	880.5
30.4	950.3	31.1	881.7
0.4	942.5	0.5	873.6
(c) $t = 40.5^\circ\text{C}$		(d) $t = 51.5^\circ\text{C}$	
0.2	733.7	0.4	631.2
5.4	735.7	5.6	633.3
10.3	737.1	10.6	634.4
15.4	738.1	15.4	635.8
20.2	740.2	20.6	637.1
25.2	742.1	25.6	639.0
30.3	743.5	31.8	640.4
0.5	734.9	0.9	631.4
(e) $t = 66.3^\circ\text{C}$		(f) $t = 92.7^\circ\text{C}$	
0.4	527.3	1.3	401.9
5.7	528.8	5.6	402.8
10.7	530.3	10.5	404.9
15.5	531.6	15.4	406.6
21.5	533.0	20.6	409.1
25.2	534.5		
32.0	535.9		
1.2	527.6		
(g) $t = 124.6^\circ\text{C}$		(h) $t = 162.1^\circ\text{C}$	
2.8	310.4	3.0	244.5
7.7	312.0	8.1	244.9
14.2	313.5	14.6	246.3
20.7	316.6	21.3	248.3
25.2	316.9	26.1	248.4
30.2	317.7	31.4	249.0
3.7	311.4	4.6	244.5
(i) $t = 204.5^\circ\text{C}$			
3.2	199.3		
8.8	200.7		
15.0	201.4		
20.7	202.5		
26.2	203.3		
32.1	205.4		
5.5	199.2		

Table V. Viscosity of Aqueous KCl (4.62550 mol · kg⁻¹)

P (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)	P (MPa)	η ($\mu\text{Pa} \cdot \text{s}$)
(a) $t = 25.4^\circ\text{C}$		(b) $t = 28.8^\circ\text{C}$	
0.3	954.4	0.2	903.9
5.3	956.6	5.3	905.1
10.3	958.0	10.7	906.9
15.4	959.3	15.7	908.4
20.5	961.5	20.7	910.8
25.1	963.5	25.7	911.6
30.5	964.6	31.0	912.9
		0.2	904.9
(c) $t = 40.6^\circ\text{C}$		(d) $t = 49.7^\circ\text{C}$	
0.3	757.0	0.2	669.1
5.4	758.5	5.5	671.0
10.6	760.1	10.6	672.5
15.9	761.7	15.8	674.2
20.8	762.9	21.3	676.1
25.9	764.9	26.5	677.9
32.0	767.6	32.7	679.8
0.4	756.8	0.2	669.5
(e) $t = 65.4^\circ\text{C}$		(f) $t = 90.5^\circ\text{C}$	
0.4	554.0	0.3	428.9
5.5	555.6	5.3	431.0
10.3	557.5	10.4	432.3
15.2	558.6	15.2	433.9
20.4	560.2	20.5	436.3
25.5	562.0	25.4	438.2
30.4	563.5	30.6	439.9
0.2	554.1		
(g) $t = 125.7^\circ\text{C}$		(h) $t = 161.0^\circ\text{C}$	
1.8	321.4	3.4	258.3
7.0	322.8	8.2	260.5
10.9	323.9	13.6	260.9
15.6	325.9	20.6	263.3
20.4	326.7	25.2	264.1
25.3	327.9	31.1	264.8
30.1	330.2	4.4	259.3
4.2	322.1		
(i) $t = 201.4^\circ\text{C}$			
3.6	213.4		
8.4	215.0		
15.3	215.3		
20.3	217.0		
25.3	217.6		
31.3	219.6		
4.6	214.4		

information, a piecewise Arrhenius fit over a limited range of temperature was employed to estimate $(\partial\eta/\partial t)_P$. As these corrections amount to less than 1% of viscosity, a reasonable estimation of $(\partial\eta/\partial t)_P$ introduces an insignificant uncertainty to the viscosity.

Measurements of the viscosity were performed from near saturation pressure to a pressure of 30 MPa at approximately 5 MPa intervals. For each isotherm, the pressure dependence of viscosity was correlated by means of the linear relationship

$$\eta(P, t, m) = \eta^\circ(t, m)[1 + \beta(t, m)P], \quad (2)$$

in which $\eta^\circ(t, m)$ denotes the hypothetical zero-pressure viscosity for a solution of molality, m , at temperature, t ; $\beta(t, m)$ denotes the viscosity-pressure coefficient for the particular concentration and temperature. The worst-case standard deviation of this linear fit represents 0.4% of η° . Over the pressure considered here, the magnitude of the change in the viscosity with pressure is relatively small ($< 4\%$). An empirical polynomial expression of the form

$$\beta(t, m) = \sum_{i=0}^3 \sum_{j=0}^3 b_{ij} t^i m^j, \quad (3)$$

sufficed to correlate the pressure coefficient as a function of temperature and concentration. The optimum values of b_{ij} are listed in Table 6; for $m = 0$, this equation reproduces the corresponding data for pure water [1].

Unfortunately, the single existing theoretical result which describes the composition dependence of the viscosity of ionic solutions is valid only at extreme dilution; this well-known relationship is of the form [12]

$$\eta_R = 1 + Ac^{1/2}, \quad (4)$$

in which η_R denotes the relative viscosity of the solution and c is the concentration (molarity) of the solution. The coefficient A can be calcu-

Table VI. Coefficients b_{ij} for Eq. (3)^a

i	$j = 0$	$j = 1$	$j = 2$	$j = 3$
0	-7.829×10^{-1}	7.704×10^{-1}	-2.832×10^{-1}	3.324×10^{-2}
1	2.927×10^{-2}	-2.904×10^{-2}	1.262×10^{-2}	-1.533×10^{-3}
2	-1.584×10^{-4}	3.470×10^{-4}	-1.627×10^{-4}	1.992×10^{-5}
3	3.775×10^{-7}	-1.265×10^{-6}	5.913×10^{-7}	-7.149×10^{-8}

^aThe b_{ij} possess units $(\text{GPa})^{-1} \cdot (\text{mol}^{-1} \cdot \text{kg})^j \cdot (\text{°C})^{-i}$.

lated from the ionic conductances and the viscosity and dielectric properties of the solvent. For aqueous KCl solutions, the $Ac^{1/2}$ term contributes less than 2% to the relative viscosity [13]. An extension of the description to wider concentration ranges would require the introduction of empirical modifications to Eq. (4). The most commonly employed extension is due to Jones and Dole [14] and can be expressed as

$$\eta_R = 1 + Ac^{1/2} + Bc. \quad (5)$$

The B coefficient is generally thought to arise from ion-solvent interactions [13] and at present is not theoretically calculable. For most electrolyte solutions, Eq. (5) requires still more empirical terms in order to describe the complete composition dependence; these most often consist of simple polynomial extensions. In the present case, we also need to correlate the temperature dependence of η° and for this reason we have chosen to develop a relatively simple correlation in terms of the molality. We have also omitted the square-root term owing to its small magnitude. The experimental data were best described by

$$\eta_R(t, m) = 1 + d_1m + d_2m^2 + d_3m^3, \quad (6a)$$

where

$$d_i = \sum_{j=0}^3 d_{ij}t^j. \quad (6b)$$

The coefficients, d_{ij} , are summarized in Table 7. The relative viscosity was computed with the aid of the correlations for the viscosity of water reported by Kestin et al. [1, 15]. Equation (6) reproduces the $\eta^\circ(t, m)$ data within an average absolute deviation of 0.2%; the individual deviations are depicted in Fig. 1. The complete data set in the temperature-pressure-concentration space is reproduced by Eqs. (2), (3), and (6) within an average absolute deviation of 0.2% and a maximum absolute deviation of 0.8%.

Table VII. Coefficients d_{ij} for Eq. (6)^a

j	$i = 1$	$i = 2$	$i = 3$
0	-8.526×10^{-2}	2.506×10^{-2}	-2.295×10^{-3}
1	3.575×10^{-3}	-9.018×10^{-4}	1.037×10^{-4}
2	-2.504×10^{-5}	9.016×10^{-6}	-1.109×10^{-6}
3	6.372×10^{-8}	-2.664×10^{-8}	3.397×10^{-9}

^aThe d_{ij} possess units $(\text{mol}^{-1} \cdot \text{kg})^i \cdot (^\circ\text{C})^{-j}$.

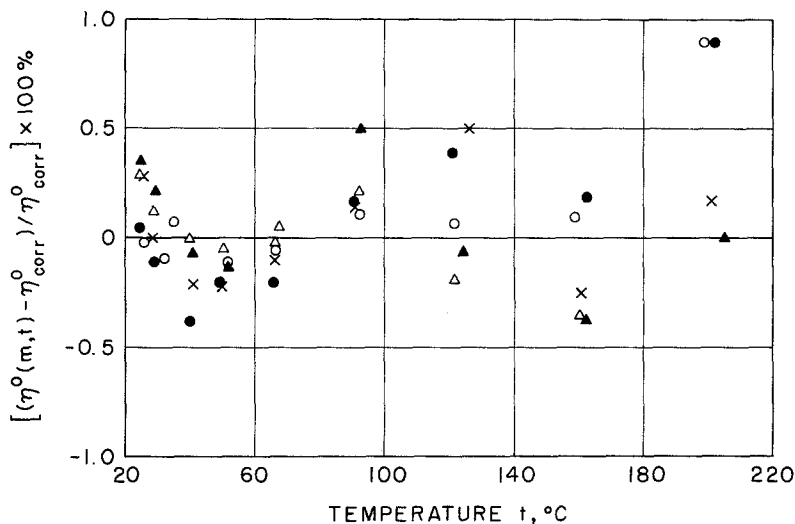


Fig. 1. Deviations between $\eta^{\circ}(t, m)$ data and the correlation represented by Eq. (6); \circ , 0.99005 mol \cdot kg $^{-1}$; \triangle , 2.07510 mol \cdot kg $^{-1}$; \bullet , 3.10040 mol \cdot kg $^{-1}$; \blacktriangle , 3.96850 mol \cdot kg $^{-1}$; \times , 4.62550 mol \cdot kg $^{-1}$.

Although Eq. (6) is purely empirical and differs slightly from Eq. (5), it is perhaps informative to compare the coefficients d_1 and B . The difference in concentration scales can be accounted for by

$$B(t) \approx d_1(t) / \rho_w(t), \quad (7)$$

where ρ_w is the density of water. This relationship is only approximate; first, because of the omission of the $m^{1/2}$ term in Eq. (6). Secondly, a good statistical estimation of d_1 (or B) would have to be based on a larger number of intermediate concentrations than made available in this study. However, inspection of Fig. 2 reveals a quite reasonable agreement, albeit over a small temperature range, between d_1/ρ_w and the previously reported B data [13, 16]. Hence, the present data may serve as a guide to the temperature dependence of the coefficient B over a larger range of temperature. Also shown in Fig. 2 is a similar comparison for our earlier results with the NaCl system [1].

Attempts were also made to fit the present data to previously employed empirical correlating equations. Korosi and Fabuss [10] measured the viscosity of aqueous KCl solutions and correlated their data with an Othmer-rule type equation, *viz.*,

$$\ln \eta_R(t, m) = A'(m) + B'(m) \ln [\eta_w(t) / \eta_w(20^\circ\text{C})], \quad (8)$$

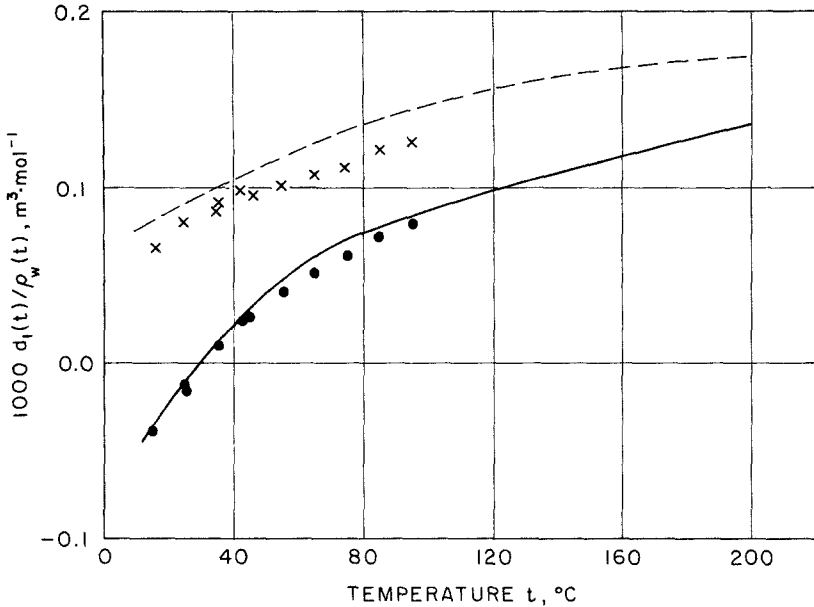


Fig. 2. Comparison of $d_1(t)/\rho_w(t)$ with the viscosity B coefficient. —, Eq. (6b); ●, viscosity B coefficient for KCl [13, 16]; ---, $d_1(t)/\rho_w(t)$ for NaCl [1]; ×, viscosity B coefficient for NaCl [13, 16].

where η_w is the viscosity of water. This form of equation correlated our earlier NaCl results [1] reasonably well, but it reproduced the present data poorly, with individual deviations as great as 3%. Arrhenius-Andrade equations, which have been used to describe the temperature dependence of the viscosity of molten salts and some electrolyte solutions [13], also failed to correlate the present data over the temperature range of measurement, although they could be used to describe the temperature dependence of $\eta^\circ(t, m)$ over a more restricted temperature range.

4. EXPERIMENTAL ACCURACY

In order to assess the accuracy of the present data, we have performed a comparison with three other studies of the KCl system. As representative of the large body of high-precision data obtained with capillary-type viscometers, we have chosen the work of Goncalves and Kestin [11]. Although this work covered a limited temperature range (20–50 $^\circ\text{C}$) at ambient pressures, it did encompass the wide concentration range 0–4.6 $\text{mol} \cdot \text{kg}^{-1}$ with an estimated accuracy of $\pm 0.1\%$. Figure 3 illustrates the deviations between the present $\eta^\circ(t, m)$ data and the correlation supplied

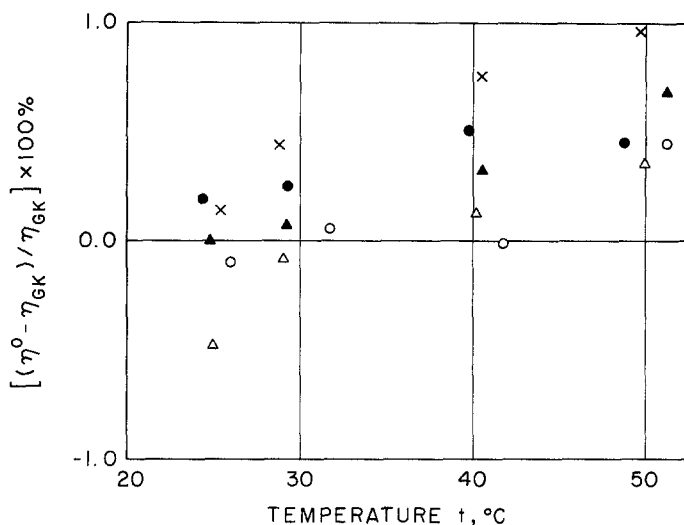


Fig. 3. Deviations between the present $\eta^0(t, m)$ data and the correlation of Goncalves and Kestin [11]; \circ , $0.99005 \text{ mol} \cdot \text{kg}^{-1}$; \triangle , $2.07510 \text{ mol} \cdot \text{kg}^{-1}$; \bullet , $3.10040 \text{ mol} \cdot \text{kg}^{-1}$; \blacktriangle , $3.96850 \text{ mol} \cdot \text{kg}^{-1}$; \times , $4.62550 \text{ mol} \cdot \text{kg}^{-1}$.

by Goncalves and Kestin. The average deviation is 0.3% while the maximum deviation is 1%. Although these discrepancies appear to be systematic, they do reflect a marked improvement upon the data obtained in this laboratory with an earlier version of the oscillating-disk viscometer [5]. Korosi and Fabuss [10] have reported viscosity data for aqueous KCl solutions over the temperature range 25–150°C and the concentration range 0–3.5 mol · kg⁻¹. Figure 4 depicts the deviations between the present data and the correlation of Korosi and Fabuss, which represents their data to within $\pm 0.5\%$. The present results deviate by an average of 1% and are systematically higher than this correlation. It is also significant to note that the form of the correlating equation employed by Korosi and Fabuss (Eq. 8) did not adequately describe the present data. The deviations between the values of $\eta^0(t, m)$ reported here and the corresponding data reported by Grimes et al. [5] are displayed in Fig. 5. Except for temperatures below 40°C, where it has been noted [5] that the data of Grimes et al. do not agree particularly well with the precise capillary data, the deviations are typically 1%.

The precision or reproducibility of the present values is $\pm 0.1\%$ near room temperature, degrading to $\pm 0.5\%$ at 200°C. In previous studies [1, 2], which have employed the current viscometer, an accuracy comparable with this precision has been claimed. However, in the present case, the results are burdened with an additional uncertainty arising from inaccuracies in

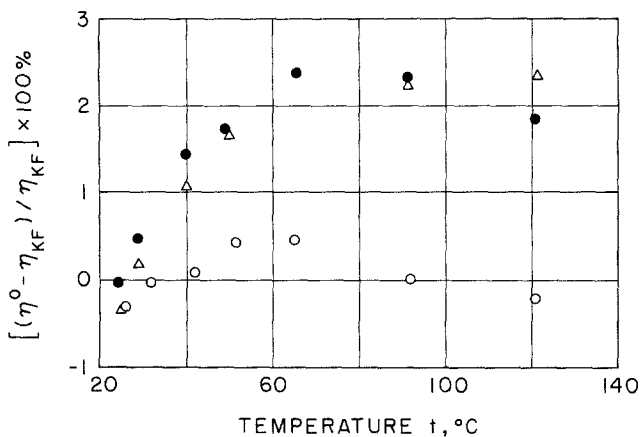


Fig. 4. Deviations between the present $\eta^{\circ}(t, m)$ data and the correlation of Korosi and Fabuss [10]; ○, 0.99005 mol · kg⁻¹; △, 2.07510 mol · kg⁻¹; ●, 3.10040 mol · kg⁻¹.

the values of density available to us. The sensitivity of the viscometer working equation to uncertainties in ρ has been evaluated numerically; it points to an error amplification effect at the highest temperatures reported here; at room temperature, the viscosity is insensitive to any uncertainty in ρ , whereas at 200°C, the relative uncertainty in ρ is amplified by a factor of

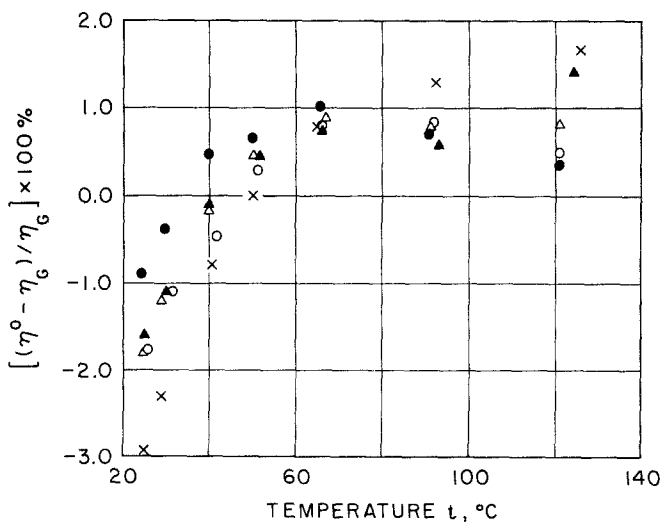


Fig. 5. Deviations between the present $\eta^{\circ}(t, m)$ data and the correlation of Grimes et al. [5]; ○, 0.990050 mol · kg⁻¹; △, 2.07510 mol · kg⁻¹; ●, 3.10040 mol · kg⁻¹; ▲, 3.96850 mol · kg⁻¹; ×, 4.62550 mol · kg⁻¹.

approximately 2 in η . As the density [5, 8, 9] could possibly be burdened with errors as great as $\pm 0.5\%$, we estimate our viscosity data to be accurate to ± 0.5 – 1.5% .

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