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Experimental Study of the Effect of Temperature, Pressure, and Concentration on the Viscosity of Aqueous $SrCl_2$ Solutions in the Range of (293 to 473) K and (0.1 to 20) MPa

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The viscosity of five [(0.27, 0.58, 0.92, 1.73, and 2.63) mol·kg^{-1}] binary aqueous $SrCl_2$ solutions has been measured with a capillary flow technique. Measurements were made at pressures up to 20 MPa. The range of temperature was (293 to 473) K. The total uncertainties in viscosity, pressure, temperature, and composition measurements were estimated to be less than 1.6 %, 0.05 %, 15 mK, and 0.02 %, respectively. The effects of temperature, pressure, and concentration on the viscosity of $SrCl_2$(aq) solutions were studied. The temperature and pressure coefficients of the viscosity of $SrCl_2$(aq) were studied as a function of concentration and temperature. The measured values of viscosity were compared with data, predictions, and correlations reported in the literature. The viscosity data were used to accurately calculate the physical meaning parameters (viscosity *A* and *B* coefficients) in the extended Jones−Dole equation for the relative viscosity (η/η_0). Various theoretical models [absolute rate theory, TTG (Tammann−Tait−Gibson) model, extended Einstein model] were used to accurately represent the measured values of viscosity. The values of hydrodynamic molar volume, V_k (effective rigid molar volume of salt), were calculated using the present experimental viscosity data. The high-pressure viscosity measurements were used to test the predictive capability of the TTG model.

Introduction

Viscosity is one of the key transport properties of electrolyte solutions. The design of engineering systems utilizing aqueous electrolyte solutions requires an accurate knowledge of transport properties (viscosity and thermal conductivity). In many industrial and natural processes, aqueous solutions are present at high temperatures and high pressures, for example, in deep geological formations (underground waters), in steam-power generation, geothermal power plants, hydrothermal synthesis, seawater desalination processes, and other industrial operations. Accurate viscosity data are known to give valuable information regarding ion−solvent interactions (long-range electrostatic interactions), which are the controlling forces in dilute solutions where ion−ion interactions are absent. Therefore, precise viscosity data are needed to accurately calculate the physical meaning parameters (for example viscosity *A* and *B* coefficients in the extended Jones−Dole equation, *V* and *E* in the Eyring's absolute rate theory, or the values of effective rigid molar volume of salt V_k in the extended Einstein model) in the various theories of electrolyte solutions. Because of severe high temperature and high-pressure conditions in aqueous systems, there are no experimental viscosity data for the $SrCl_2$(aq) solutions at high temperatures and high pressures.

Literature Review of the Experimental Data and Correlations. A survey of the literature reveals a scarcity of experimental viscosity data for $SrCl_2$(aq) solutions, especially at high temperatures and high pressures. Only nine data sources were found for the viscosity of $SrCl_2$(aq)[1−9] at the atmospheric pressure and at temperatures up to 328.15 K. Afzal et al.[1] reported viscosities for $SrCl_2$(aq) solutions in the temperature range from (293 to 323) K at atmospheric pressure. Measurements were made with a Ubbelohde viscometer. The results of the measurements were described with the equation

$$\eta = a_0 \exp(b_0 c + c_0 c^2) \qquad (1)$$

where *c* is the electrolyte concentration (mol·L^{-1}); and a_0, b_0, and c_0 are temperature-dependent coefficients. Nayaka et al.[2] measured the viscosity of dilute $SrCl_2$(aq) solutions (0.05 and 0.1 mol·kg^{-1}) at temperatures from (303.15 and 318.15) K. Herz[3] reported viscosities of $SrCl_2$(aq) solutions at 298.15 K and 0.1 MPa. Isono[4,5] reported viscosities for $SrCl_2$(aq) solutions in the temperature range from (288 to 328) K and concentrations from (0.05 to 2.5) mol·kg^{-1} at atmospheric pressure. Measurements were made with a Ubbelohde-type viscometer. No uncertainty in the viscosity measurements is available for the source data. The measured values of viscosity were used to calculate the temperature coefficient $(1/\eta)(\partial\eta/\partial T)_{P,X}$ as a function of concentration at a temperature of 298.15 K. The measured values of viscosity were fitted to the extended Jones−Dole type equation

$$\eta/\eta_0 = 1 + A\sqrt{m} + Bm + Dm^2 + Em^3 \qquad (2)$$

where η and η_0 are the viscosities of an electrolyte solution and pure solvent (water), respectively; *A* is always a positive constant, and *m* is the electrolyte concentration (mol·kg^{-1}). The coefficients *B*, *D*, and *E* are the polynomial type functions of temperature. This equation is valid in the temperature range from (288 to 328) K.

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Table 1. Test Measurements of Viscosity of Pure Water at Atmospheric Pressure

T/K	this work	IAPWS[29]	$\delta\eta^a$/%
293.15	0.9986	1.0020	−0.34
298.15	0.8921	0.8901	+0.22
313.15	0.6551	0.6530	+0.32
343.15	0.4053	0.4039	−0.35
363.15	0.3128	0.3144	−0.51
365.15	0.3055	0.3070	−0.49
AAD		0.37 %	

[a] $\delta\eta = 100(\eta^{\exp} - \eta^{\mathrm{IAPWS}})/\eta^{\exp}$.

Table 2. Experimental Values of the Viscosities, Pressures, Temperatures, and Molalities of $SrCl_2$(aq) Solutions

T	η/mPa·s		
K	$P = 0.1$ MPa	$P = 10$ MPa	$P = 20$ MPa
$m = 0.27$ mol·kg^{-1}			
293.15	1.0797	1.0790	1.0789
298.15	0.9628	0.9634	0.9639
313.56	0.7071	0.7094	0.7108
347.50	0.4209	0.4246	0.4271
363.86	0.3461	0.3510	0.3552
399.45		0.2482	0.2509
425.88		0.2030	0.2075
450.35		0.1733	0.1761
469.45		0.1558	0.1586
$m = 0.58$ mol·kg^{-1}			
293.15	1.1720	1.1715	1.1694
298.15	1.0475	1.0468	1.0482
313.90	0.7701	0.7775	0.7876
348.57	0.4614	0.4654	0.4676
364.49	0.3822	0.3857	0.3890
397.85		0.2809	0.2841
424.40		0.2301	0.2333
448.96		0.1957	0.1988
471.50		0.1721	0.1758
$m = 0.92$ mol·kg^{-1}			
293.15	1.2831	1.2830	1.2824
298.15	1.1518	1.1521	1.1526
315.86	0.8261	0.8286	0.8308
349.57	0.5150	0.5164	0.5186
362.54	0.4472	0.4492	0.4510
398.99		0.3198	0.3240
425.50		0.2623	0.2661
447.39		0.2275	0.2303
471.27		0.1955	0.1994
$m = 1.73$ mol·kg^{-1}			
293.15	1.6066	1.6070	1.6082
298.15	1.4472	1.4477	1.4489
313.20	1.0960	1.0981	1.0993
348.47	0.6649	0.6694	0.6746
364.39	0.5664	0.5701	0.5766
399.86		0.4168	0.4217
424.53		0.3497	0.3547
450.00		0.2967	0.3018
472.54		0.2600	0.2648
$m = 2.63$ mol·kg^{-1}			
293.15	2.0653	2.0667	2.0674
298.15	1.8619	1.8624	1.8636
314.54	1.3744	1.3769	1.3798
347.43	0.8724	0.8757	0.8795
363.87	0.7243	0.7311	0.7372
398.96		0.5314	0.5374
424.51		0.4444	0.4506
445.39		0.3924	0.3980
472.98		0.3348	0.3409

Zaytsev and Aseyev[6] represented available experimental viscosity data from the literature[2,7,8] for aqueous $SrCl_2$(aq) solutions in the temperature range from (273 to 373) K and at concentrations from (5 to 60) mass % at atmospheric pressure by the following correlation equation:

$$\log_{10}\eta = \log_{10}\eta_0 + 10^{-2}(0.979 + 0.00196t)x \tag{3}$$

where η_0 is the viscosity of pure water in (mPa·s), x is the composition in (mass %), and t is the temperature in (°C). The absolute uncertainty of calculated values of the viscosity of $SrCl_2$(aq) is 0.0214 Pa·s. The TTG (Tammann−Tait−Gibson) model for the viscosity of $SrCl_2$(aq) solutions at 293 K were reported by Leyendekkers[9] and Leyendekkers and Hunter.[10] The developed model fitted the viscosity data within the experimental uncertainty (0.3 to 0.5 %) at low concentrations and about 2 % at high concentrations. Only two adjustable parameters are needed to accurately represent the experimental data using this model over a wide concentration range (from dilute to saturation). This model can also be used to predict the viscosity of electrolyte solutions at high pressures. The compilation of the volumetric and transport properties of $SrCl_2$(aq) at atmospheric pressure have also been reported by Chapman and Newman.[11] This compilation includes only the viscosity data reported by Timmermans[12] at 298.15 K and 0.1 MPa.

It is apparent that experimental viscosity data at high temperatures (above 328 K) and high pressures (above 0.1 MPa) for $SrCl_2$(aq) solutions are not available in the literature. Thus, the primary objective of this work was to expand the existing viscosity database for $SrCl_2$(aq) to higher temperatures (up to 473 K), pressures (up to 20 MPa), and concentrations (up to 2.63 mol·kg^{-1}). The main objective of the paper is to provide new accurate experimental viscosity data for binary $SrCl_2$(aq) solutions at high temperatures and high pressures using a capillary-flow technique. This technique has been previously used for accurate viscosity measurements of other aqueous salt solutions.[13−27] This work is part of a continuing program on the transport (thermal conductivity and viscosity) and thermodynamic ($P-V-T-x$) property measurements of electrolytes in aqueous solutions at high temperatures and high pressures. In previous studies,[13−27] we measured the viscosity of 17 aqueous salts (Na_2SO_4, K_2SO_4, Li_2SO_4, $MgSO_4$, Na_2CO_3, $MgCl_2$, LiCl, $BaCl_2$, $CaCl_2$, $NaNO_3$, NaBr, $Ca(NO_3)_2$, $LiNO_3$, $Ni(NO_3)_2$, $BaNO_3$, LiI, NaI) solutions at high temperatures (up to 607 K) and high pressures (up to 60 MPa) by using the capillary-flow technique. The values of the viscosity of concentrated $SrCl_2$(aq) solutions covering the range of temperatures and pressures reported in this work are not available in the literature.

Experimental Section

Since this method (apparatus, procedure, and uncertainty assessment) has been described fully in our other papers,[13−27] it will be not be reproduced here. The measurements were made using a capillary-flow method, which gives an uncertainty of 1.6 % for the viscosity. The technique is based on Poiseuille's law, which relates viscosity to the rate of fluid flow through a capillary tube. The main parts of the apparatus consisted of a working capillary with an extension tube, a high temperature and high pressure autoclave, movable and unmovable cylinders, electrical heaters, and solid red copper block. The capillary together with the extension tube was located in the high temperature and high pressure autoclave. When the movable cylinder was moved vertically at constant speed, the fluid flowed through the capillary. The autoclave was placed in the solid red copper block. To generate and measure the pressure, the autoclave was connected with a dead-weight pressure gauge (MP-600) by means of a separating vessel. The maximum uncertainty in pressure measurements was 0.05 %.

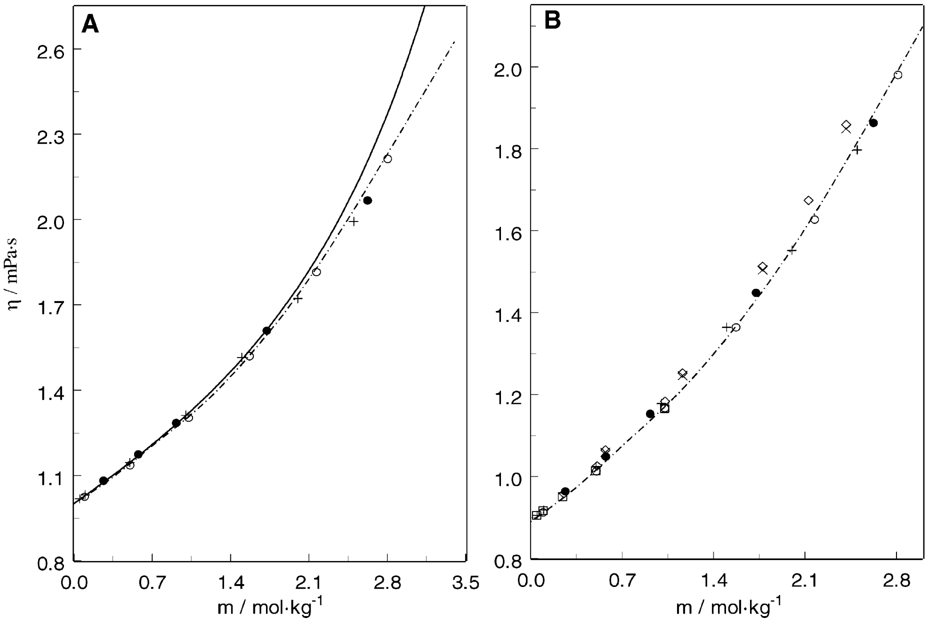

Figure 1. Measured values of viscosity η of $SrCl_2$(aq) solutions as a function of concentration *m* along two selected isotherms of 293.15 (A) and 298.15 (B) and at atmospheric pressure together with published data. Panel A: ○, ref 1; ●, this work; +, refs 4 and 5; —, TTD model by Leyendekkers;[9,10] – • –, correlation by Zaytsev and Aseyev.[6] Panel B: ○, ref 1; ●, this work; +, refs 4 and 5; ◇, ref 12; ×, ref 3; □, ref 12; – • –, correlation by Zaytsev and Aseyev.[6]

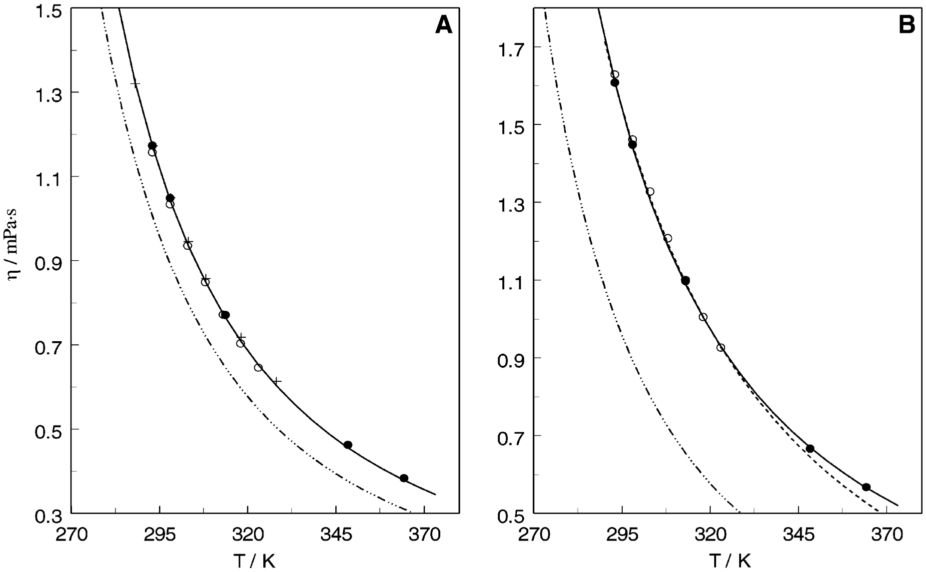

Figure 2. Measured values of viscosity η of $SrCl_2$(aq) solutions as a function of temperature *T* at two selected concentrations of 0.58 (A) and 1.73 (B) $mol\cdot kg^{-1}$ and at atmospheric pressure together with reported data. ○, ref 1; ●, this work; +, (exp) refs 4 and 5; – –, correlation by Isono;[4] —, ref 6; ··· —···, pure water (IAPWS, ref 29).

The time for fluid to flow through the capillary τ was measured with a stopwatch with an uncertainty of less than 0.1 s (0.5 %). Based on the detailed analysis of all sources of uncertainties likely to affect the determination of viscosity with the present apparatus, the combined maximum relative uncertainty $\delta\eta/\eta$ in measuring the viscosity was 1.6 % (see ref 13). The Reynolds (*Re*) number occurring during all measurements was (about 300) less than the critical value of 2000 for this method.

To calculate the dynamic viscosity from measured quantities, the values of density of the solution under study at room temperature and experimental pressure and the density at the experimental conditions $\rho(P,T)$ are needed. For this purpose, we used the density data, $\rho(P,T)$, reported in our previous work.[28]

As a check of the method and procedure of the measurements, before engaging in measurements on aqueous $SrCl_2$(aq) solutions, we measured the viscosity of triple-distilled water at atmospheric pressure in the temperature range from (293 to 365) K. The results are given in Table 1 along with values from the IAPWS formulation[29] for pure water. The differences are very small (AAD = 0.37 %) and are less than their experimental uncertainty (1.6 %). This good agreement between the present data and the IAPWS[29] calculations for pure water confirms the reliability and high accuracy of the measurements for $SrCl_2$(aq) solutions and gives us an assurance that our apparatus is functioning correctly.

The $SrCl_2$(aq) solutions were prepared from chemically pure $SrCl_2$ (Merck GR, > 99.5 mass %) and triple-distilled water. The solutions were prepared gravimetrically with an analytical

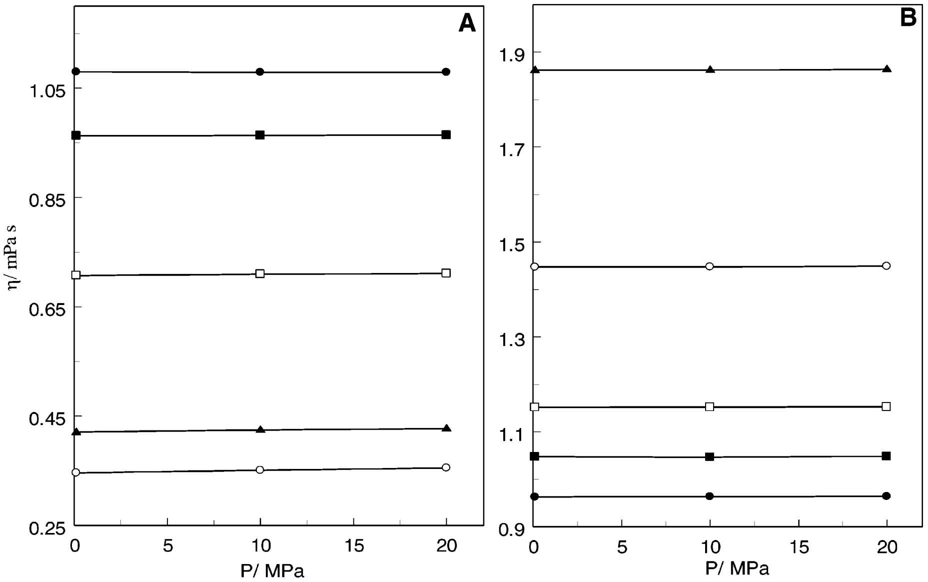

Figure 3. Measured values of viscosity η of $SrCl_2$(aq) solutions as a function of pressure P at selected isotherms (A, $m = 0.27$ mol·kg^{-1}) and isopleths (B, $T = 298.15$ K). Panel A: ●, 293.15 K; ■, 298.15 K; □, 313.56 K; ▲, 347.50 K; and ○, 363.15 K. Panel B: ●, 0.27 mol·kg^{-1}; ■, 0.58 mol·kg^{-1}; □, 0.92 mol·kg^{-1}; ○, 1.73 mol·kg^{-1}; and ▲, 2.63 mol·kg^{-1}. The solid lines guide the eye.

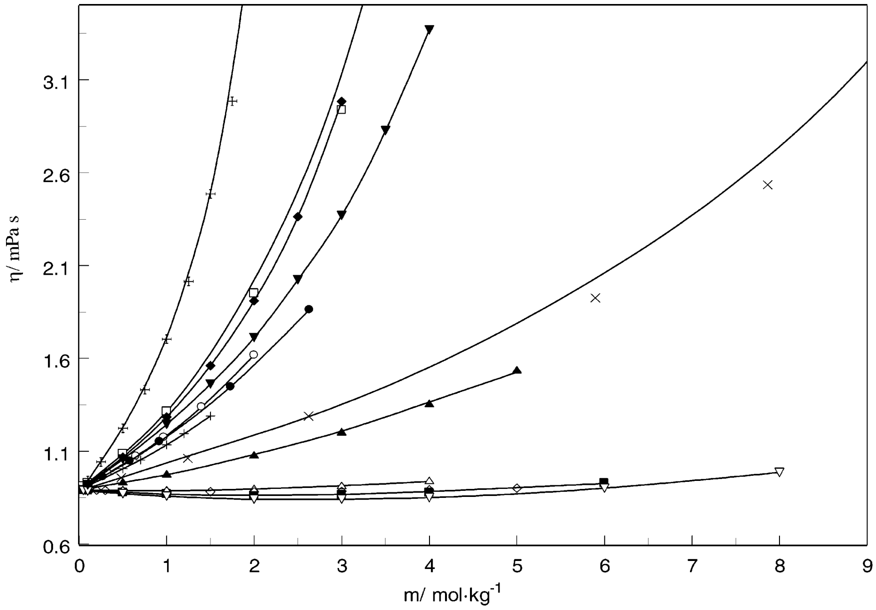

Figure 4. Viscosity of a series of aqueous solutions as a function of composition at a selected temperature 298.15 K and a pressure of 0.1 MPa reported by various authors. ●, $SrCl_2$ (this work); □, $MgCl_2$ (ref 26); ×, LiCl (ref 23); ○, $CaCl_2$ (ref 29); ◇, NH_4Cl (refs 4 and 5); △, KCl (ref 54); ▲, NaCl (ref 54); ■, RbCl (ref 54); ▽, CsCl (ref 54); +, $BaCl_2$ (refs 4 and 5); ◆, $CoCl_2$ (ref 1); ▼, $CuCl_2$ (ref 1); and solid cross, $CrCl_3$ (ref 1).

balance having a precision of $\pm 5 \times 10^{-8}$ kg. The solutions at the desired composition were prepared by carefully weighing the appropriate mass. To check the accuracy we determined the density of each solution at room temperature (298.15 K) and atmospheric pressure with the aid of a pycnometer accurate to 0.005 % and compared it with the reference data.

Results and Discussion

The viscosity for five [(0.27, 0.58, 0.92, 1.73, and 2.63) mol·kg^{-1}] binary aqueous $SrCl_2$ solutions has been measured in the temperature range of (293 to 473) K at pressures up to 20 MPa. All experimental viscosity data were obtained as a function of temperature at three isobars (0.1, 10, and 20 MPa). The experimental temperature, viscosity, pressure, and composition values for the aqueous $SrCl_2$ solutions are presented in Table 2. Some selected experimental viscosity data for $SrCl_2$(aq) solutions as an example of the present results are shown in Figures 1 to 3 in the $\eta-m$, $\eta-T$, and $\eta-P$ projections together with values reported by other authors. These figures also include the values of viscosity of $SrCl_2$(aq) calculated with the correlations by Afzal et al.,[1] Isono,[5] Zaytsev and Aseyev,[6] and Leyendekkers.[9,10]

Figure 4 demonstrates the concentration dependence of the viscosity of a series of aqueous solutions with the same anion (Cl^-) and various cations (K^+, Na^+, Li^+, Rb^+, Cs^+, NH_4^+, Sr^{2+}, Ba^{2+}, Mg^{2+}, Co^{2+}, Cu^{2+}, Ca^{2+}, and Cr^{3+}) reported by various authors. This figure demonstrates the effect of the nature of the solute ions (cations) on the values and concentration dependence behavior of the viscosity of aqueous salt solutions (chlorides). As one can see from Figure 4, the $SrCl_2$(aq) solutions showed the intermediate values of viscosity among other aqueous solutions at the same thermodynamic (P, T, m) conditions. From this figure, it is also apparent that the values and concentration dependence of the viscosity of aqueous electrolyte solutions

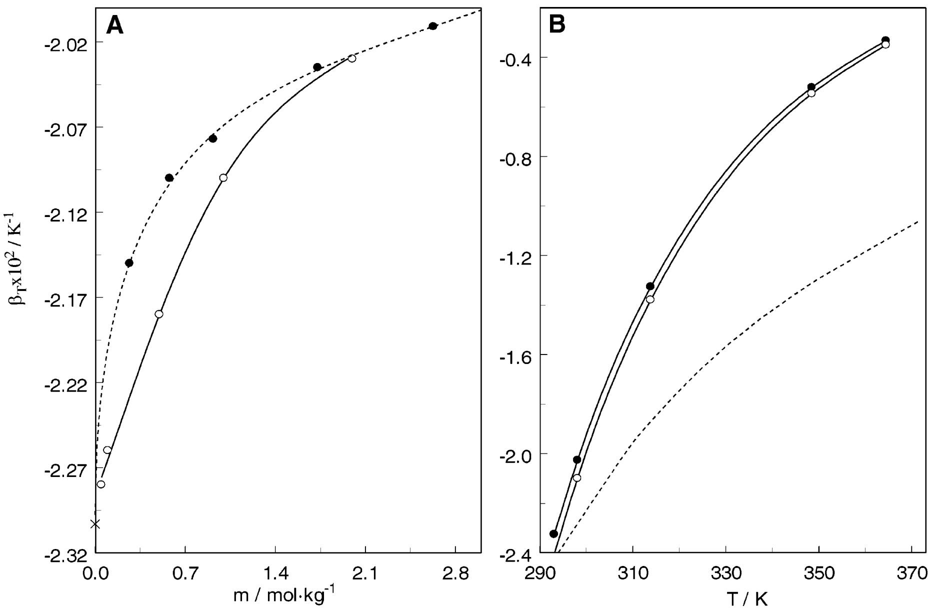

Figure 5. Temperature coefficient, β_T, of $SrCl_2$(aq) solution and pure water as a function of concentration (A, $T = 298.15$ K) and temperature (B) at atmospheric pressure. Panel A: ●, this work; ○, ref 5; ×, pure water (IAPWS, ref 29); − −, smoothed curve. Panel B: ●, 0.92 mol·kg^{-1}; ○, 0.27 mol·kg^{-1}; − −, pure water (IAPWS, ref 29); —, smoothed curves.

Table 3. Values of Parameters ln *A*, *b*, and *c* (eqs 4 and 5) for $SrCl_2$(aq) as a Function of Composition at Atmospheric Pressure

m	eq 4		eq 5		
mol·kg^{-1}	ln *A*	*b*/K	ln *A*	*b*/K	$c \times 10^5$/K^2
0.27	−5.7884	1714.18	−1.8547	−857.88	4.173638
0.58	−5.5679	1673.66	−1.8737	−744.31	3.927036
0.92	−5.2718	1613.51	−1.1038	−1108.2	4.411264
1.73	−4.8939	1568.74	−0.7011	−1175.7	4.457298
2.63	−4.6694	1576.84	−1.1252	−739.91	3.758538

Table 4. Values of Parameters *V*, *E*, and Viscosity *B* Coefficient (eqs 7 and 8) for $SrCl_2$(aq) as a Function of Temperature

T/K	*V*	*E*	*B*/L·mol^{-1}
293.15	11.451	26.0	0.2621
298.15	10.199	25.5	0.2756
313.56	0.1486	18.0	0.3216
347.50	0.2111	19.5	0.3475
363.86	0.2438	20.0	0.3559

(chlorides) considerably depends on ionic valence, shape, and size of the cations.

Comparison with Other Data and Correlations. The present experimental values for the viscosity for $SrCl_2$(aq) solutions were compared with data reported by other authors at atmospheric pressure. Figures 1 and 2 demonstrate the direct comparison of the present viscosity data for $SrCl_2$(aq) solutions with those reported by various authors in the literature. As one can see from these figures, basically the agreement between the various data sets is good, except the two data sets reported by Herz[3] and Timmermans.[12] These data are systematically higher (about 6 to 9 %) than the present and other published data. The relative percentage deviations between the present viscosity data for $SrCl_2$(aq) solutions and the data reported by other authors are within 1.2 % at low concentrations, while at high concentrations the deviations are up to 2 %. Majority of the reported data at atmospheric pressure and low temperatures lie between (0.5 and 1.7) %, which is close to the experimental estimated uncertainty (1.6 %). Excellent agreement, within 0.11 %, is found between the present measurements and the data reported by Isono[4,5] at a temperature of 293.15 K and at atmospheric pressure. The measurements by Afzal et al.[1] agree with the present results within (0.5 and 1.8) % at low concentrations (below 2.5 mol·kg^{-1}). Excellent agreement within (0.15 to 0.58) % is also found between the present data and the values calculated with the Leyendekkers[9,10] TTG model for the viscosity of electrolyte solutions (see Figure 1A) at concentrations up to 1.73 mol·kg^{-1}. At high concentrations, deviations reached up to (5 to 6) %. As Leyendekkers[9,10] claimed, the uncertainty of calculated values of viscosity at high concentrations is within 2 %. The values of viscosity calculated with the correlation by Zaytsev and Aseyev[6] agree with the present results within (0.4 to 0.6) % at atmospheric pressure and at low temperatures and low concentrations (up to 1.73 mol·kg^{-1}). At high concentrations and high temperatures, the deviations reach up to 2.5 %.

Temperature Dependence of the Viscosity of $SrCl_2$(aq) Solutions. The selected experimental viscosity data for $SrCl_2$(aq) solutions as a function of temperature are shown in Figure 2 in the $\eta - T$ plane along two constant concentrations (0.58 and 1.73) mol·kg^{-1}. The viscosity of the solution decreases considerably with temperature. The temperature coefficients of viscosity, $\beta_T = \eta^{-1}(\partial\eta/\partial T)_{P,m}$, for aqueous $SrCl_2$ solutions calculated with the present experimental data are depicted in Figure 5 as a function of concentration and temperature. This figure also includes the values of β_T reported by Isono[4] for $SrCl_2$(aq). As one can see from Figure 5A, the temperature coefficient, β_T, rapidly increases with concentration at $m < 1.5$ mol·kg^{-1}. At high concentrations, β_T changes only slightly with concentration. As was shown in our previous paper[30] and by Isono,[4] for NaBr(aq) the temperature coefficient of viscosity passes through a maximum near 4 mol·kg^{-1} and then decreases at higher concentrations. For $SrCl_2$(aq) the concentration range where the maximum in β_T should be observed was not reached, although the concentration behavior of β_T at high concentrations most likely confirms the earlier observation. Figure 5B also demonstrates the temperature dependence of β_T at 0.1 MPa for various fixed concentrations of $SrCl_2$(aq) and pure water calculated with the IAPWS[29] formulation. As this figure shows, the effect of temperature on the viscosity of $SrCl_2$(aq) solutions is higher than that of pure water and monotonically increases with temperature.

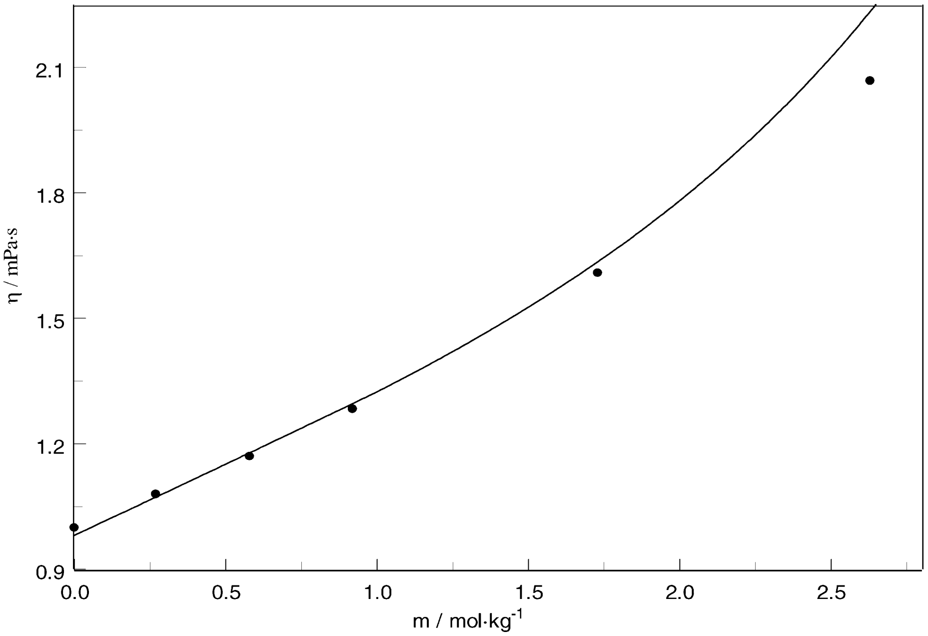

Figure 6. Measured and predicted values of the viscosity of $SrCl_2$(aq) as a function of concentration at pressure of 20 MPa and at 293.15 K. ●, this work; —, predicted by TTG model (eq 10) of Leyendekkers.[9,10]

In the concentration range $m > 1$ mol·kg $^{-1}$, the empirical equation of Arrhenius−Andrade is valid for the temperature dependence of viscosity:[31−33]

$$\eta = A \exp(b/T) \qquad (4)$$

where A and b are functions of concentration. The values of these parameters for $SrCl_2$(aq) solutions calculated using the present measurements at atmospheric pressure as a function of concentration are given in Table 3. This equation reproduces the present viscosity data within 0.85 % in the temperature range from (293 to 364) K. However, eq 4 failed to represent the present data over the complete temperature range at high pressures. One more term

$$\eta = A \exp(b/T + c/T^2) \qquad (5)$$

is required to accurately describe the experimental viscosity data at high temperatures. The values of the parameters A, b, and c are also given in Table 3. The Eyring's absolute rate theory[33] enables a detailed description of the temperature dependence of viscosity of concentrated aqueous electrolyte solutions:

$$\eta = \frac{hN}{V} \exp\left(\frac{\Delta G^*}{RT}\right) \text{ or } \eta = A \exp\left(\frac{\Delta H^*}{RT}\right) \qquad (6)$$

where ΔG^* and ΔH^* are the molar free energy of activation and enthalpy of activation, respectively, h is Planck's constant, N is Avogadro's constant, R is the gas constant, and V is the molar volume of the hole in the liquid. The enthalpy of activation $\Delta H^*/R$ (see Table 3 where the value of $b = \Delta H^*/R$) can be calculated from the slope of the straight line by the ln $\eta \approx 1/T$ function. In simplified form, eq 5 can be given by[34,35]

$$\eta = \frac{\eta_0 \exp(xE)}{1 + xV} \qquad (7)$$

where η is the viscosity of the electrolyte solution at a concentration m and temperature T, η_0 is the viscosity of the solvent (pure water) at temperature T, x is the mole fraction of the cation or anion in solution, and the parameters E and V are defined as $E = (\Delta G_c^* + \Delta G_a^* - 2\Delta G_0^*)/RT$ and $V = ((V_c + V_a)/V_0) - 2$, where V_c is the molar volume of the cation particle, V_a is the molar volume of the anion particle, V_0 is the molar volume of the solvent particle (pure water), ΔG_c^* is the molar free energy of activation for viscous flow of the cation particle, ΔG_a^* is the molar free energy of activation for viscous flow of the anion particle, ΔG_0^* is the molar free energy of activation for viscous flow of the solvent particle, and $x = m/(55.5 + 2m)$, where m is the concentration of the salt in mol·kg^{-1}. The temperature dependence of the viscosity of concentrated aqueous electrolyte solutions can be explained in terms of the temperature dependence of the E and V parameters in eq 7 for the viscosity of solutions. The results of the application of eq 7 to the present viscosity measurements for $SrCl_2$(aq) are presented in Table 4 as a function of temperature. The Jones−Dole viscosity B coefficient (see below) is related to the parameters V and E by the relation

$$B = (E - V)/55.51 \qquad (8)$$

The values of the viscosity B coefficients calculated with eq 8 using the values of V and E parameters for $SrCl_2$(aq) are given in Table 4.

Pressure Dependence of the Viscosity of $SrCl_2$(aq) Solutions. The viscosity is affected marginally by pressure along the isotherms-isopleths (see Figure 3). The pressure dependence of the experimental viscosity of $SrCl_2$(aq) in the range from (0.1 to 20) MPa is almost linear.

Leyendekkers[9] and Leyendekkers and Hunter[10] have applied the TTG model to calculate the viscosity of aqueous electrolyte solutions at high pressures. According to the TTG model the viscosity equation can be represented by

$$\eta_S = \eta_{wis} \exp(a_1 m + a_2 m^2 + ...) \qquad (9)$$

where η_S and η_{wis} represent the viscosities of the solution and the water in solution, respectively, and m is the concentration

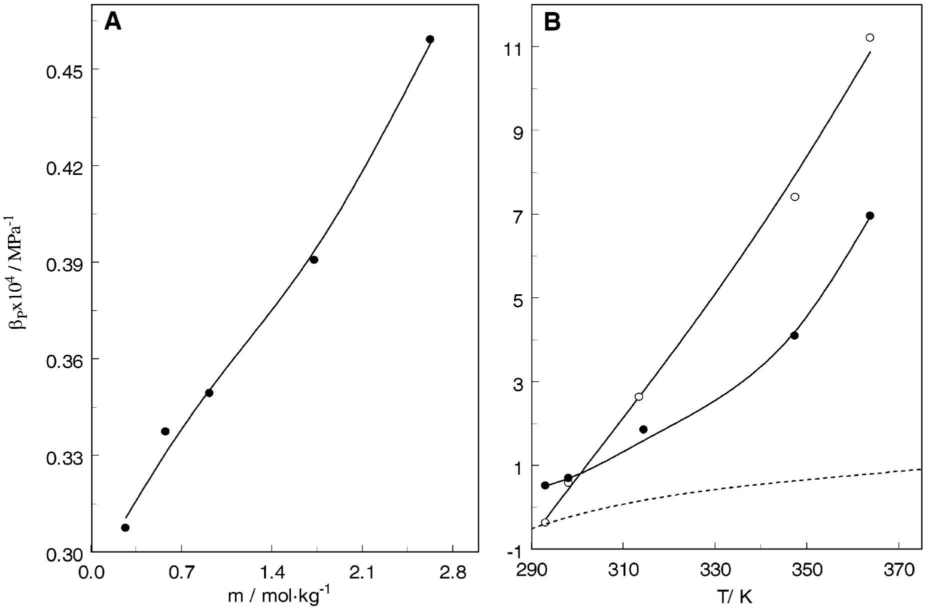

Figure 7. Pressure coefficient, β_P, of $SrCl_2$(aq) and pure water as a function of concentration (A, $T = 298.15$ K) and temperature (B) at atmospheric pressure. Panel A: ●, this work; —, smoothed curves. Panel B: ●, 0.27 mol·kg^{-1}; ○, 2.63 mol·kg^{-1}; − −, pure water (IAPWS, ref 29); —, smoothed curves.

(in mol·kg^{-1}). This model predicts the viscosity of the solution at pressure P as

$$\frac{\eta_S^{(P)}}{\eta_S^{(1)}} = \frac{\eta_W^{(P_e+P)}}{\eta_W^{(P_e+1)}} \tag{10}$$

where $\eta_W^{(P_e+P)}$ is the viscosity of pure water at pressure ($P_e + P$), $\eta_S^{(P)}$ is the viscosity of solution at pressure P, $\eta_S^{(1)}$ is the viscosity of solution at 1 bar, P is the external pressure, and P_e is the effective pressure due to the salt:[10]

$$P_e = h_0 + h_1x + h_2x^2 + h_3x^3 \tag{11}$$

where x is the concentration of the solute in grams per gram of solution and the h_i parameters that are functions of temperature only. We applied eq 9 to the $SrCl_2$(aq) data in order to calculate the viscosity at high pressures. The calculations were performed using the P_e from eq 11 of Leyendekkers and Hunter[10] and the IAPWS[29] formulation for pure water at a temperature of 293.15 K. Figure 6 shows the comparison between the predicted and the present measured values of viscosity of $SrCl_2$(aq) at a pressure of 20 MPa and at a temperature of 293.15 K. The agreement between the measured and predicted values of viscosity at concentrations up to 1.5 mol·kg^{-1} is within (0.2 to 0.6) %, while at high concentration it is within (5 to 6) %. The lack of the P_e data as a function of temperature precluded the testing of eq 9 over the full temperature range of the measurements.

The present data were used to calculate pressure coefficients, $\beta_P = \eta^{-1}(\partial\eta/\partial P)_{T,m}$, of viscosity for $SrCl_2$(aq) solutions at 0.1 MPa. The derived values of the pressure coefficient, β_P, of the solution as a function of concentration and temperature together with values for pure water calculated with the IAPWS[29] formulation are presented in Figure 7. As one can see from this figure, the pressure coefficient of solution is higher than pure water values and negative at low temperatures. The expected uncertainty in the pressure coefficients, β_P, of viscosity is about (10 to 15) %.

Concentration Dependence of the Viscosity of $SrCl_2$(aq) Solutions. The concentration dependence of the viscosity of electrolyte solutions is anomalous. For some electrolyte solutions such as $H_2O + NaCl$, $H_2O + LiCl$, $H_2O + LiBr$, $H_2O + LiI$, $H_2O + NaF$, $H_2O + KF$, $H_2O + RbF$, and $H_2O + CsF$, the viscosity increases monotonically with the electrolyte concentration, while for other types of electrolyte solutions such as $H_2O + KCl$, $H_2O + RbCl$, $H_2O + CsCl$, $H_2O + KI$, and $H_2O + KBr$, the viscosity decreases with concentration at low electrolyte concentrations reaching a minimum value and then increases monotonically at higher concentrations (see Figure 4). As one can see from Figures 1, 2, and 4, the viscosity of $SrCl_2$(aq) solutions monotonically increases with the electrolyte concentration without a minimum.

Table 5. Viscosity *A*, *B*, and *D* Coefficients (eq 13) of $SrCl_2$(aq) Solutions as a Function of Temperature at Atmospheric Pressure

T/K	A/L$^{1/2}$·mol$^{-1/2}$	B/L·mol^{-1}	D/L^2·mol^{-2}
293.15	0.0158	0.2197	0.0881
298.15	0.0291	0.2259	0.0791
313.56	0.0397	0.2491	0.0879
347.50	0.0450	0.3202	0.0827
363.86	0.0460	0.3534	0.0769

The existing theoretical result, which describes the concentration dependence of the viscosity of ionic solutions, is valid for only very dilute solutions. The Falkenhagen−Onsager−Fuoss[36,37] and Debye−Hückel−Onsager[38,39] theory predicts a square root concentration dependence of the viscosity of ionic solutions at infinite dilution, $\eta/\eta_0 \propto \sqrt{c}$ ($c \to 0$). This theory correctly explains the rise of viscosity with concentration in the limit of very low (dilute solutions) ion concentrations ($c <$ 0.05 mol·L^{-1}). Jones and Dole[40] proposed an empirical extension of the Falkenhagen and Dole[36] model to high concentrations as

$$\eta/\eta_0 = 1 + A\sqrt{c} + Bc \tag{12}$$

for the viscosity of electrolyte solutions. In eq 12, η and η_0 are the viscosities of an electrolyte solution and pure solvent (water), respectively; A is always a positive constant, and c is the electrolyte concentration (mol·L^{-1}). Falkenhagen and Dole[36] gave a theoretical derivation of the A coefficient. According the theory, the values of A coefficient can be calculated from the values of equivalent conductances $\lambda_{\pm}^{\infty}$ at infinite dilution of the ions, the viscosity of the pure solvent (water) η_0, and relative

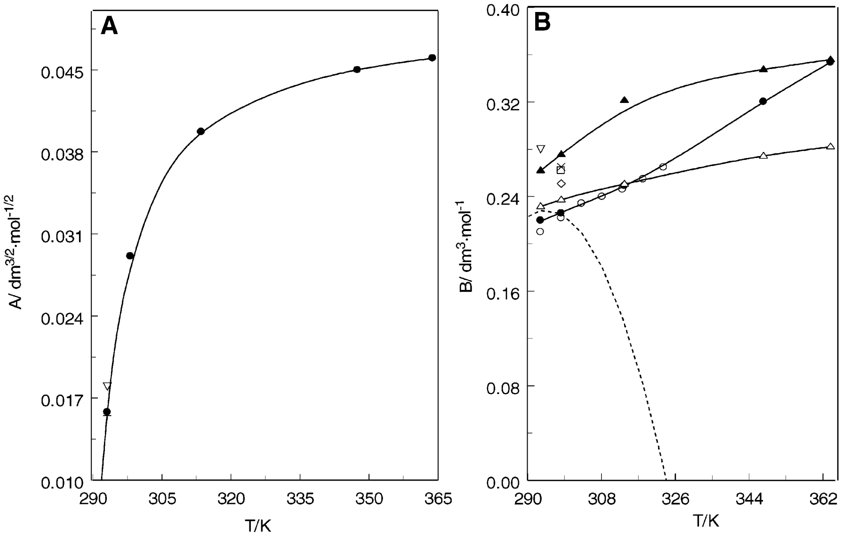

Figure 8. Experimental viscosity *A* and *B* coefficients of the $SrCl_2$(aq) solutions as a function of temperature together with values reported by other authors. △, this work from eq 15; ●, this work from eq 13; ○, ref 1; ◇, ref 41; ×, ref 55; ▲, this work from eq 8; □, ref 41; ▽, refs 9 and 10; +, ref 4; − −, ref 4.

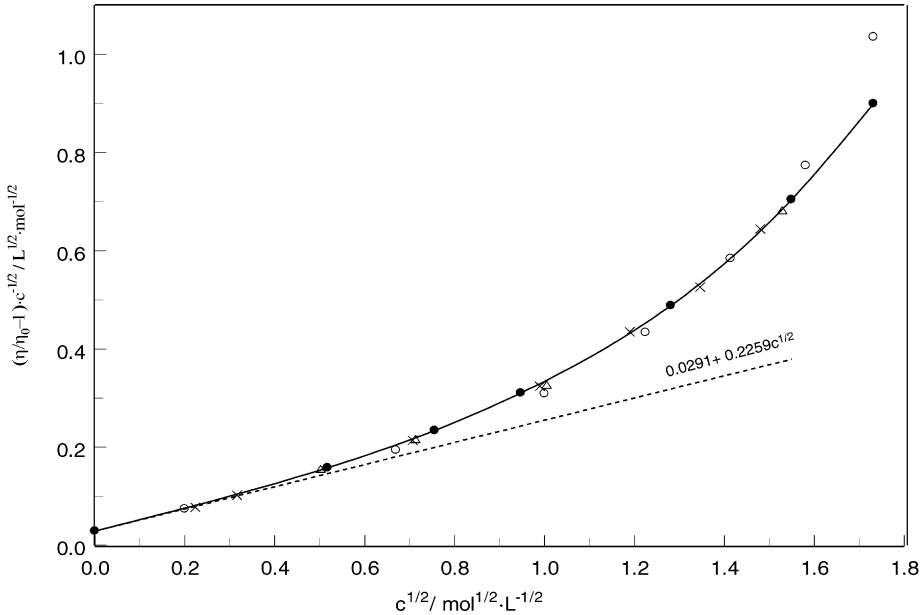

Figure 9. Jones−Dole plot, $[(\eta/\eta_0) - 1]/c^{1/2}$ vs $c^{1/2}$for $SrCl_2$(aq) solutions for selected isotherm of 298.15 at atmospheric pressure. ●, this work; ○, ref 1; ×, refs 4 and 5; △, ref 12; —, smoothed curve; − −, limiting slope.

permittivity (dielectric constant) ϵ_0 of pure water. The values of the viscosity *B* coefficient in eq 12 can be determined by adding individual contributions of the solute constituent ions $B = z^+B^- + z^-B^+$, where $z^{\pm}$ and $B^{\pm}$ refer to the ionic valence and ionic (cationic and anionic) viscosity *B* coefficients.[41−43] The values of $B^{\pm}$ are constant at a given *T* for given ions in a specific solvent and describe solely the ion−solvent interactions. The values of $B^{\pm}$ for some ions at different temperatures are available in the literature (see, for example, ref 41). The values of the viscosity *A* and *B* coefficients of an electrolyte provide information on the interaction between dissolved ions (Sr^{2+}, Cl^-) and molecules of a solvent (H_2O). The theory cannot predict the values of the viscosity *B* coefficient. Usually the *B* coefficient is considered as adjustable parameters and can be calculated by using precise experimental viscosity data. The viscosity *A* and *B* coefficients of electrolyte solutions are useful tools in the study of structural interactions (ion−ion, ion−solvent, and solvent−solvent) in solutions. An extensive compilation of Jones−Dole *A* and *B* coefficients for the series aqueous electrolyte solutions reported by various authors is available in the literature.[31,41−49] The sign of the *B* coefficient depends on the degree of solvent structuring introduced by the ions. A positive value of the *B* coefficient is associated with structure−making (ordering) ions, while a negative value of the *B* coefficient is associated with structure-breaking (disordering) ions. There are very limited (only at two temperatures (278 and 298) K) ionic viscosity *B* coefficient data for Sr^{2+} (see, for example, ref 41). For Sr^{2+} and Cl^-, the values of $dB^{\pm}/dT$ are positive, while for other ions are negative (for example, Mg^{2+} and Li^+) or almost zero (for example Na^+). Therefore, the measurements of temperature dependence for the viscosity *A* and *B* coefficients of $SrCl_2$(aq) are very limited. In the present work, we studied the viscosity *A* and *B* coefficients values of $SrCl_2$(aq) solutions as a function of temperature in the temperature range from (293 to 364) K.

The Jones−Dole eq (eq 12) has been extended by some authors[45,46,50−52] by adding a quadratic term Dc^2

$$\eta/\eta_0 = 1 + A\sqrt{c} + Bc + Dc^2 \qquad (13)$$

Table 6. Values of V_k (eq 15) for $SrCl_2(aq)$ as a Function of Temperature at Atmospheric Pressure

T/K	V_k/L·mol^{-1}	B/L·mol^{-1}
293.15	0.09265	0.2316
298.15	0.09491	0.2373
313.56	0.10021	0.2505
347.50	0.10971	0.2743
363.86	0.11286	0.2822

to apply the Jones−Dole equation for a wide concentration range. The new Dc^2 term of eq 13 includes all solute−solvent and solute−solute structural interactions that were not accounted by the $A\sqrt{c}$ and Bc terms at high concentrations such as[50] high terms of the long-range Coulombic forces; high term hydrodynamic effect; and interactions arising from changes in solute−solvent interactions with concentration. The present experimental data for the relative viscosity (η/η_0) of $SrCl_2(aq)$ for various temperatures together with data reported by other authors at low concentrations were used to calculate the values of viscosity A, B, and D coefficients in the extended Jones−Dole eq (eq 13). The results are summarized in Table 5 and presented in Figure 8 as a function of temperature together with values reported by other authors. As one can see from Figure 8, the agreement between A and B coefficients derived in the present study and calculated with ionic B coefficients data and various correlations is satisfactory, except the values calculated with the Isono[4] correlation at high temperatures. Excellent agreement is found between the present data and the values reported by Afzal et al.[1] The values of B coefficients calculated from ionic B coefficients data are systematically higher (about 0.025 dm^3·mol^{-1} or 10 %) than the present experimental data. The single data points (0.0155 and 0.0180 dm$^{3/2}$·mol$^{-1/2}$) at 293.15 K reported by Isono[4] and Leyendekkers,[9] respectively, for the viscosity A coefficient are in satisfactory agreement with the present result of 0.0158 dm$^{3/2}$·mol$^{-1/2}$. Figure 8 also demonstrates that the viscosity A coefficient monotonically increases with temperature and the derivative (temperature coefficient) $dB/dT > 0$ is positive (structure-breaking ions).

To estimate the values of the viscosity A and B coefficients, we also used the Jones−Dole plot technique (i.e., the slope of the experimental dependence $[(\eta/\eta_0) - 1]/c^{1/2}$ on $c^{1/2}$; see Figure 9):

$$[(\eta/\eta_0) - 1]/c^{1/2} = A + Bc^{1/2} \tag{14}$$

The experimental values of the viscosity A and B coefficients are determined by plotting the left side of eq 14, $[(\eta/\eta_0) - 1]/c^{1/2}$, against $c^{1/2}$. Figure 9 illustrates such plots for $SrCl_2(aq)$ solutions at a selected temperature of 298.15 K. The intercepts and slopes of the linear plots are the viscosity A and B coefficients, respectively.

The extended Einstein relation for the relative viscosity[53]

$$\eta/\eta_0 = 1 + 2.5\phi + 10.05\phi^2 = 1 + 2.5V_kc + 10.05{V_k}^2c^2 \tag{15}$$

has been used to represent the present viscosity data for $SrCl_2(aq)$. In eq 15, ϕ is the volume fraction of the solute molecules ($\phi = 4/3\pi R^3N_Ac$, where R is the effective solute ions radius or hydrodynamic radius of the ions and c is the salt concentration), V_k is the hydrodynamic molar volume in cm^3·mol^{-1}. The B coefficient is related to molar volume V_k by $B = 2.5V_k$. Equation 15 was applied to the present relative viscosity data for $SrCl_2(aq)$, where V_k was taken as an adjustable parameter. The derived values of V_k are given in Table 6. Equation 15 fitted the present data within the experimental uncertainty in the whole experimental concentration range. As one can see only one physically meaning parameter V_k is need to accurately represent the measured values of the viscosity of $SrCl_2(aq)$. Therefore, eq 15 can be used to accurately estimate the values of the hydrodynamic volume V_k by using the experimental relative viscosity data. The values of the viscosity B coefficients derived from eq 15 are illustrated also in Figure 8. As one can see from Figure 8, the agreement between the viscosity B coefficients derived from eq 15 are in good agreement with the values calculated from eq 13, while the data calculated with eq 8 are systematically higher.

Conclusions

The values of the viscosity A, B, and D coefficients of the extended Jones−Dole equation for the relative viscosity (η/η_0) as a function of temperature were derived from the precise viscosity measurements for $SrCl_2(aq)$. The physical meaning parameters V and E in the absolute rate theory of viscosity and hydrodynamic molar volumeV_k in the extended Einstein relation were calculated using the present experimental viscosity data as a function of temperature. The temperature (β_T) and pressure (β_P) coefficients of viscosity for $SrCl_2(aq)$ are calculated using the present data. The effect of temperature on the viscosity of the $SrCl_2(aq)$ solutions is higher than that of pure water. From the data obtained, it can be concluded that the pressure coefficient β_P for the solution and water is negative at low temperatures (below 300 K) and positive at high temperatures. It was found that the TTG model is suitable to predict the values of viscosity of $SrCl_2(aq)$ solutions at high pressures. It was also shown that the extended Einstein relation for the relative viscosity with only one physical meaning parameter V_k (hydrodynamic molar volume or effective rigid molar volume of salt) can be used to accurately represent the present viscosity data for $SrCl_2(aq)$ as a function of concentration over a wide concentration range.

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