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Viscosity of Aqueous Electrolyte Solutions at High Temperatures and High Pressures. Viscosity *B*-coefficient. Sodium Iodide

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The viscosity of seven (0.062, 0.166, 0.456, 0.552, 1.754, 3.062, and 4.700) mol·kg^{-1} binary aqueous NaI solutions has been measured with a capillary-flow technique. Measurements were performed at pressures up to 30 MPa. The range of temperature was from (298 to 575) K. The total uncertainty of viscosity, pressure, temperature, and composition measurements was estimated to be less than 1.6 %, 0.05 %, 15 mK, and 0.015 %, respectively. The effect of temperature, pressure, and concentration on viscosity of binary aqueous NaI solutions was studied. The measured values of the viscosity of NaI(aq) were compared with data, predictions, and correlations reported in the literature. The viscosity data have been analyzed and interpreted in terms of the extended Jones−Dole equation for the relative viscosity (η/η_0) of strong electrolyte solutions to accurately calculate the values of the viscosity *A*- and *B*-coefficients as a function of temperature. The derived values of the viscosity *A*- and *B*-coefficients were compared with the values calculated from the Falkenhagen−Dole theory and ionic *B*-coefficient data, respectively. The physical meaning parameters V and E in the Eyring's absolute rate theory of viscosity, the hydrodynamic molar volume V_k (effective rigid molar volume of salt) in the extended Einstein relation for the relative viscosity, and the Arrhenius−Andrade parameters A and $b = E_a/R$ (where E_a is the flow activation energy) were calculated using the present experimental viscosity data. The effective pressures P_e due to the salt (NaI) in water were calculated from the present viscosity measurements by using the TTG model. The predictive capability of the various models for viscosity electrolyte solutions has been tested.

Introduction

Transport properties (viscosity, electrical and thermal conductivities, diffusivity) of aqueous electrolyte solutions in a wide range of solute concentrations and solution temperatures and pressures are of fundamental importance for the understanding of various physicochemical processes occurring in the chemical industry and in the natural environment, for example, crystallization, osmotic desalination, wastewater treatment, pollution control, oil recovery, heat and mass transfer, fluid flow, mineral transport and deposition, corrosion, electrometrical energy storage devices, portable energy systems (rechargeable batteries, lithium-ion cells), electrochemical technology, and so forth.[1−3] In many applications (chemical engineering and geochemistry), these processes occur at high temperatures and high pressures. Aqueous solutions are present at high temperature and high pressure in deep geological formations. Aqueous systems at high temperatures also arise in steam-power generation, geothermal power plants, hydrothermal synthesis, seawater desalination processes, and other industrial operations. The oceans and undergroundwaters are the largest reservoirs of aqueous electrolyte solutions at high temperatures and high pressures. Groundwaters encountered in deep wells drilled in crystalline rocks also commonly are highly saline brines in which Na^+ is the dominant cation. Na^+ is one of the dominant cations (most important cation) of many aqueous fluids (natural fluids). Thus, there is great practical interest in the thermodynamic and transport properties of aqueous salt solutions at high temperatures and high pressures.

The viscosity of electrolyte solutions is also of research interest because of the long-range electrostatic interactions (Coulombic forces) between ions.[4−14] Temperature and concentration dependencies of the viscosity of aqueous electrolytes solutions are crucial also for understanding ion−solvent (solvation) and long-range ion−ion electrostatic interactions. Accurate viscosity data are needed to calculate the physical meaning of the viscosity *A*- and *B*-coefficients in the limiting law of viscosity and extended Jones−Dole viscosity equation, respectively. Furthermore, the Jones−Dole viscosity *B*-coefficient is important in the description and understanding of the structure and destruction of ionic processes, for example, solvation effects of cations and anions.[4−14] The variations of the solvational properties are reflected in the viscosity *B*-coefficients of ions. The theory predicts only the values of the viscosity *A*-coefficient of electrolyte solutions at infinite dilution ($m \rightarrow 0$) (Falkenhagen and Dole[15]). To accurately determine the higher-degree viscosity coefficients (e.g., *B*- and *D*- coefficients) in the extended Jones−Dole equation, reliable and very accurate viscosity data for electrolyte solutions at a high concentration range are required. Theoretical modeling[8−13,16−20] of the viscosity of electrolyte solutions still cannot accurately predict the behavior of real systems in wide temperature, pressure, and concentrations ranges. To develop better predictive models for electrolyte solutions, more reliable viscosity measurements in wider temperature, pressure, and concentration ranges are required.

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Table 1. Summary of the Viscosity Measurements for Aqueous NaI Solutions

first author	method[a]	uncertainty %	temperature K	pressure MPa	concentration $mol \cdot kg^{-1}$
Goldsack[21,22]	OV	n.a.	298−328	0.1	0.5−3.0
Kacperska,[29] Taniewska-Osinska[30,31]	Ub	0.02	288−313	0.1	0.27−1.07
Sahu[27]	OV	n.a.	298	0.1	0.097−3.48
Lengyel[24]	OV	n.a.	283−313	0.1	1.1472−11.286
Satoh[23]	OV	n.a.	296	0.1	0.485−4.35
Desnoyers[25]	Ub	0.01	298	0.1	0.0254−0.6808
Dunlop[26]	OV	3.6	298	0.1	0.29−2.65
ICT[32]		n.a.	283−333	0.1	0.5−8.0
Ebert[28]	Ub	n.a.	293−363	0.1	8.0
This work	CV	1.6	298−575	0.1−30	0.062−4.700

[a] CV: capillary viscometer; OV: Ostwald-type viscometer; Ub: Ubbelohde-type viscometer; n.a.: no uncertainty given in source reference.

Literature Review. Experiment. A literature survey revealed that the number of measurements reported for the viscosity of aqueous NaI(aq) at atmospheric pressure cover very limited ranges of temperature (up to 363 K) and concentration. In Table 1, a summary of all viscosity measurements for NaI(aq) is presented. A survey of the literature reveals the lack of reliable experimental viscosity data for NaI(aq) solutions at high temperatures and high pressures (see Table 1). Furthermore, the scatter of the reported data at atmospheric pressure is quite large (up to 20−30 %, especially at high concentrations) and exceeds the quoted mutual uncertainties of the authors. Here, the published data are briefly reviewed. Most reported experimental data on the viscosity of NaI(aq) cover near ambient temperatures (298 to 363) K and atmospheric pressure. Most of the data were obtained using a Ubbelohde and Ostwald-type viscometer. Some of the reported viscosity data are inaccurate and inconsistent. Goldsack and Franchetto[21,22] reported viscosities of aqueous NaI solutions in the temperature range from (298 to 328) K and at atmospheric pressure for concentrations between (0.5 and 3.0) $mol \cdot kg^{-1}$. The measurements were made with an Ostwald-type microviscometer. The measured values of viscosity were used to calculate the values of the parameters V and E in the absolute rate theory of viscosity. The temperature dependence of the viscosity of concentrated aqueous NaI solutions has been explained in terms of the temperature dependence of the V and E parameters. Satoh and Hayashi[23] reported viscosities of NaI(aq) solutions at a temperature of 296 K and at concentrations from (0.485 to 4.35) $mol \cdot L^{-1}$. Measurements were made with an Ostwald-type viscometer. Lengyel et al.[24] also measured the viscosity of NaI(aq) with an Ostwald-type viscometer for concentrations from (1.1472 to 11.286) $mol \cdot kg^{-1}$ at temperatures between (283 and 313) K. Desnoyers and Perron[25] reported the viscosity data for dilute NaI(aq) solutions in the concentration range from (0.0254 to 0.6808) $mol \cdot L^{-1}$ and at 298 K to accurately determine the viscosity A-, B-, and D-coefficients. Measurements were performed with a Ubbelohde-type viscometer. Dunlop[26] reported viscosity data for NaI(aq) at 298 K. The uncertainty of the measured values of viscosity is 3.6 %. Measurements were made with an Ostwald-type viscometer. Sahu and Behera[27] employed an Ostwald-type viscometer to measure the viscosity of NaI(aq) solutions at 298 K and in the concentration range from (0.097 to 3.48) $mol \cdot kg^{-1}$. The measured data were used to calculate the values of the Jones−Dole B-coefficient and the effective rigid molar volume V_k in the Einstein relation at 298 K. Ebert and Wendorff[28] measured the viscosity of NaI(aq) with a Ubbelohde-type viscometer in the temperature range from (293 to 363) K and at concentrations up to 8 $mol \cdot L^{-1}$. The measured results were presented graphically. Kacperska et al.[29] and Taniewska-Osinska and Kacperska[30,31] employed a Ubbelohde-type viscometer to measure the viscosity of NaI(aq) solutions at temperatures between (288 and 313) K and at concentrations from (0.27 to 1.07) $mol \cdot kg^{-1}$. The authors claimed that the uncertainty of the derived values of relative viscosity was about 0.02 %. The reported viscosity data were used to calculate the viscosity A- and B-coefficients in the Jones−Dole equation at 298 K. Zaytsev and Aseyev[33] presented available experimental viscosity data for aqueous NaI solutions in the temperature range from (273 to 373) K and at concentrations from (0.74 to 11.4) $mol \cdot kg^{-1}$ at atmospheric pressure by the following correlation equation

$$\log_{10} \eta = \log_{10} \eta_0 + (0.0064 + 0.0000558t)x \tag{1}$$

where η_0 is the viscosity of pure water in mPa·s, x is the composition in mass fraction, and t is the temperature in °C. The absolute uncertainty of the calculated values of the viscosity of NaI was 0.072 Pa·s.

It is apparent that experimental viscosity data at high temperature and high pressure for aqueous NaI solutions are not available. Thus, the primary objective of this work was to expand the existing viscosity database to higher temperatures, pressures, and concentrations. The main objective of the paper is to provide new, accurate experimental viscosity data for binary NaI(aq) solutions at high temperatures (up to 575 K) and high pressures (up to 30 MPa) for compositions up to 4.7 $mol \cdot kg^{-1}$ using a capillary-flow technique, which has been previously used for accurate measurements on other aqueous electrolyte solutions at high temperatures and high pressures.[34−47] The available experimental viscosity data for NaI(aq) have been comprehensively analyzed to estimate the reliability and consistency of the published data sets. The present results considerably expand the temperature, pressure, and concentration ranges in which viscosity for aqueous NaI(aq) are available. This work is a part of a continuing program[34−47] on the transport (thermal conductivity and viscosity) properties of electrolytes in aqueous solutions at high temperatures and high pressures.

Modeling. Jiang and Sandler[12] proposed a new equation to describe the concentration dependence of the viscosity of various types of electrolyte solutions based on the combination of liquid-state theory and absolute-rate theory. The adjustable parameters in the model ($\sigma_1^+ = 0.0953$, $\epsilon_1 = 0.0728$, $b = -0.1393$ for NaI(aq)) have physical meaning and are related to the degree of ion hydration. This model was used to describe the concentration dependence of viscosity for NaI(aq) solutions at atmospheric pressure and 298.15 K. The average relative deviation is about 2.28 % at concentrations up to 7.85 $mol \cdot L^{-1}$. Esteves et al.[10] also developed a new model for correlating the viscosity of binary strong electrolyte solutions. The proposed model was based on Eyring's absolute rate theory and the Debye−Hückel model for calculating the excess (electrostatic)

Table 2. Test Measurements of the Viscosity (mPa·s) of Pure Water

	$P = 10$ MPa		$P = 30$ MPa	
T/K	this work	IAPWS[57]	this work	IAPWS[57]
310.85	0.6849	0.6828	0.6883	0.6848
345.28	0.3964	0.3949	0.4028	0.4002
387.18	0.2459	0.2477	0.2512	0.2529
423.95	0.1851	0.1838	0.1875	0.1887
486.04	0.1270	0.1278	0.1335	0.1325
543.84	0.0985	0.0985	0.1051	0.1040
deviation statistics	AAD = 0.58 %; Bias = 0.14 %; St.Dev = 0.65 %; St.Err = 0.19 %; Max.Dev = 1.05 %			

free energy of activation of the viscous flow. The two adjustable parameters $A = 0.0136$ and $B = 1.0279$ of the model for NaI(aq) have been determined using the reported data. This model was used to correlate experimental viscosity data for NaI(aq) solutions at 298.15 K and 0.1 MPa in the concentration range from (0.0254 to 2.947) mol·kg^{-1}. The overall average mean relative standard deviation (MRSD) is 0.05 %.

Experimental Apparatus and Procedures

The details of the experimental method, the description of the apparatus, and the procedures used for the viscosity measurements of NaI(aq) solutions are given in several previous publications.[34−43] Only a brief review will be given here. The measurements were made using a capillary-flow method. This method is most frequently used for the measuring of the viscosity of aqueous electrolyte solutions at high temperatures and high pressures (see, for example, refs 48−53). 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 a solid red copper block. The working capillary with an i.d. (inner diameter) of 0.3 mm and length of 216 mm is made from stainless steel. The typical value for the capillary radius is 0.15091 mm. The value of the capillary length is 540.324 ± 0.005 mm. To create and accurately measure the pressure, the autoclave is connected to a dead-weight pressure gauge (MP-600) by means of a separating vessel. Flow time measurements were made electronically with an uncertainty of less than 0.1 s (0.5 %). The Reynolds (Re) number occurring during all measurements was less than the critical values ($Re_c = 2000$). To calculate the dynamic viscosity from measured quantities, the densities of the solution $\rho(P,T)$ are needed. For this purpose, we used the density data, $\rho(P,T)$, reported in previous publications by Akhundov et al.[54−56] for NaI(aq) solutions. After a very careful analysis of the uncertainty of all of the measured quantities (Abdulagatov and Azizov[34]), it is estimated that the combined relative uncertainty in measuring the viscosity (up to the highest temperature of 575 K) is 1.6 %.

For testing the performance of the apparatus, we have measured the viscosity of triple-distilled pure water. The measured values of the viscosity of pure water along two selected isobars (10 and 30) MPa and over the temperature range from (311 to 544) K are presented in Table 2. These data were compared with values calculated from the IAPWS formulation (Kestin et al.[57]). The agreement between both sets of data along the isobars is excellent (AAD = 0.58 %, the maximum deviation is 1.05 %). This excellent agreement confirms the reliability and high accuracy of the measurements for NaI(aq) solutions and gives us an assurance that our instrument is operating correctly.

The NaI(aq) solutions were prepared from chemically pure NaI (Merck GR, >99.5 mass %) and triple-distilled water. The solutions were prepared gravimetrically with an analytical balance having a precision of $\pm 5 \times 10^{-8}$ kg. The solutions at the desired composition were prepared by mass by careful weighing. 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 measurements for seven (0.062, 0.166, 0.456, 0.552, 1.754, 3.062, and 4.700) mol·kg^{-1} binary aqueous NaI solutions have been performed in the temperature range from (298 to 575) K at pressures up to 30 MPa. All experimental viscosity data were obtained as a function of temperature at three isobars (0.1, 10, and 30) MPa. The experimental temperature, viscosity, pressure, and composition values for the aqueous NaI solutions are presented in Table 3. Some selected experimental viscosity data for NaI(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 NaI(aq) calculated with the correlations of Zaytsev and Aseyev[33] and of Goldsack and Franchetto.[21,22]

Comparison with Other Data and Correlation. The present experimental values of the viscosity for NaI(aq) solutions at atmospheric pressure were compared with the data and correlations reported by other authors in the literature (Figures 1 to 3). As one can see from these figures, basically the agreement between various data sets at low concentrations is good (0.2−0.9 % at 298 K and 1.2−1.4 % at 313 K), except for some scattering (up to 20−30 %)[27,32] at high concentrations ($m > 3$ mol·kg^{-1}). These figures illustrate that our data are consistent with most literature values at various temperatures and concentrations. This good agreement between the present and published data confirms the reliability of the present measurements and its consistency with literature data.

Concentration Dependence of the Viscosity of NaI(aq) Solutions. For some aqueous 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, then increases monotonically for higher concentrations (see also Figure 9 below). As one can see from Figure 1, the viscosity of NaI(aq) solutions increases monotonically very slightly with the electrolyte concentration without a minimum at low concentrations (up to 1.5 mol·kg^{-1}), then increases rapidly at high concentrations.

The existing theoretical result which describes the concentration dependence of the viscosity of ionic solutions is valid only at very dilute concentrations ($c \to 0$). Falkenhagen−Onsager−Fuoss[15,58] and Debye−Hückel−Onsager[59,60] theories predict a square root concentration dependence, $\eta_r \propto c^{1/2}$, of the relative viscosity ($\eta_r = \eta/\eta_0$, where η_0 is the viscosity of pure water) of ionic solutions at infinite dilution ($c \to 0$). 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}). The dilute solution theories[15,58−60] of viscosity are not practicable because of their very limited concentration range. Jones and

Table 3. Experimental Viscosities, Pressures, Temperatures, and Concentrations of NaI(aq) Solutions

	η (mPa·s)		
	$P = 0.1$ MPa	$P = 10$ MPa	$P = 30$ MPa
T/K	m=0.062 mol·kg^{-1}		
298.15	0.8921		
302.42	0.8116	0.8119	0.8128
321.75	0.5635	0.5655	0.5698
344.64	0.4002	0.4028	0.4083
367.46	0.3042	0.3078	0.3125
387.38		0.2520	0.2575
408.99		0.2102	0.2154
432.31		0.1779	0.1830
455.25		0.1547	0.1590
479.90		0.1358	0.1403
504.62		0.1210	0.1258
537.90		0.1048	0.1103
574.60		0.0891	0.0960
	m = 0.166 mol·kg^{-1}		
298.15	0.8954		
311.38	0.6828	0.6837	0.6868
329.64	0.5015	0.5041	0.5094
345.42	0.4023	0.4052	0.4109
367.48	0.3106	0.3136	0.3195
387.11		0.2590	0.2646
411.36		0.2123	0.2177
430.74		0.1854	0.1906
452.55		0.1625	0.1674
469.89		0.1486	0.1535
489.55		0.1351	0.1400
504.81		0.1263	0.1310
522.52		0.1169	0.216
543.87		0.1065	0.1125
571.85		0.0939	0.1011
	m = 0.456 mol·kg^{-1}		
298.15	0.9034		
309.52	0.7161	0.7167	0.7187
330.94	0.5016	0.5046	0.5094
347.39	0.4011	0.4041	0.4098
365.45	0.3249	0.3281	0.3338
386.83		0.2657	0.2717
402.81		0.2318	0.2375
424.53		0.1976	0.2029
445.44		0.1732	0.1784
470.80		0.1510	0.1560
489.98		0.1377	0.1422
507.67		0.1270	0.1328
526.82		0.1175	0.1230
544.95		0.1090	0.1140
572.29		0.0967	0.1035
	m = 0.552 mol·kg^{-1}		
298.15	0.9076		
305.82	0.7724	0.7739	0.7742
326.55	0.5388	0.5413	0.5459
349.60	0.3926	0.3957	0.4014
365.96	0.3259	0.3290	0.3348
391.45		0.2575	0.2636
406.89		0.2268	0.2322
423.95		0.2000	0.2050
444.84		0.1753	0.1806
T/K	m = 0.552 mol·kg^{-1}		
463.41		0.1579	0.1623
484.82		0.1409	0.1462
444.84		0.1753	0.1806
463.41		0.1579	0.1623
484.82		0.1409	0.1462
497.90		0.1338	0.1384
522.75		0.1214	0.1264
547.95		0.1099	0.1162
568.45		0.1005	0.1360
	m = 1.754 mol·kg^{-1}		
298.15	0.9506		
305.85	0.8131	0.8142	0.8150
321.75	0.6170	0.6192	0.6236
332.41	0.5284	0.5314	0.5367
348.05	0.4330	0.4360	0.4436
365.45	0.3568	0.3605	0.3668
388.95		0.2880	0.2942
400.05		0.2628	0.2688
412.30		0.2404	0.2464
424.52		0.2218	0.2280
443.93		0.1972	0.2028
464.95		0.1760	0.1815
483.72		0.1612	0.1670
505.60		0.1477	0.1535
527.95		0.1359	0.1421
	m =3.062 mol·kg^{-1}		
298.15	1.0409		
308.74	0.8494	0.8512	0.8521
322.30	0.6801	0.6833	0.6881
334.90	0.5734	0.5772	0.5834
352.01	0.4682	0.4715	0.4794
368.56	0.3916	0.3956	0.4031
389.98		0.3267	0.3343
402.43		0.2967	0.3041
415.64		0.2697	0.2762
431.75		0.2444	0.2510
444.40		0.2287	0.2355
465.90		0.2040	0.2104
489.64		0.1841	0.1911
516.93		0.1658	0.1730
	m = 4.700 mol·kg^{-1}		
298.15	1.2154		
302.50	1.1145	1.1157	1.1152
318.70	0.8458	0.8489	0.8542
337.62	0.6563	0.6613	0.6688
353.74	0.5613	0.5662	0.5748
370.98	0.4756	0.4804	0.4887
391.41		0.4085	0.4179
405.10		0.3734	0.3824
423.33		0.3354	0.3440
444.44		0.3004	0.3090
461.58		0.2778	0.2864
483.79		0.2543	0.2634
501.44		0.2395	0.2491
521.55		0.2238	0.2340

Dole[61] proposed an empirical extension of the Falkenhagen and Dole[15] and Falkenhagen[62,63] model to high concentrations as

$$\eta/\eta_0 = 1 + A\sqrt{c} + Bc \tag{2}$$

for the viscosity of electrolyte solutions. In eq 2, η and η_0 are the viscosities of an electrolyte solution and pure solvent (water), respectively, A is an always positive constant, c is the electrolyte molarity concentration (mol·L^{-1}), and B is the empirical extension viscosity B-coefficient (Jones−Dole coefficient). Falkenhagen and Dole[15] gave a theoretical derivation of the A-coefficient. According to this theory, the value of parameter A can be calculated from the ionic equivalent conductance $\lambda_{\pm}^{\infty}$ at infinite dilution of the ions, the viscosity of the solvent η_0 (pure water, IAPWS[57]), its relative permittivity (dielectric constant, IAPWS[64]) ϵ_0, and the temperature T. The value of the viscosity A-coefficient of the Jones−Dole equation is determined by an ion−atmosphere interaction and ionic mobility. The values of the A-coefficient are a measure of long-range ion−ion interaction (Coulombic forces), while B-coefficients are a manifestation of ion−solvent interaction. At a given concentration, the B-coefficient can be interpreted in terms of competition between specialized viscosity effects (Coulombic interaction, size and shape of effects or Einstein effect, alignment or orientation of polar molecules by the ionic field, and distortion of the solvent structure). These effects govern the viscosity behavior of the aqueous electrolyte solutions. The values of the viscosity B-coefficient are a highly specific property of the solute

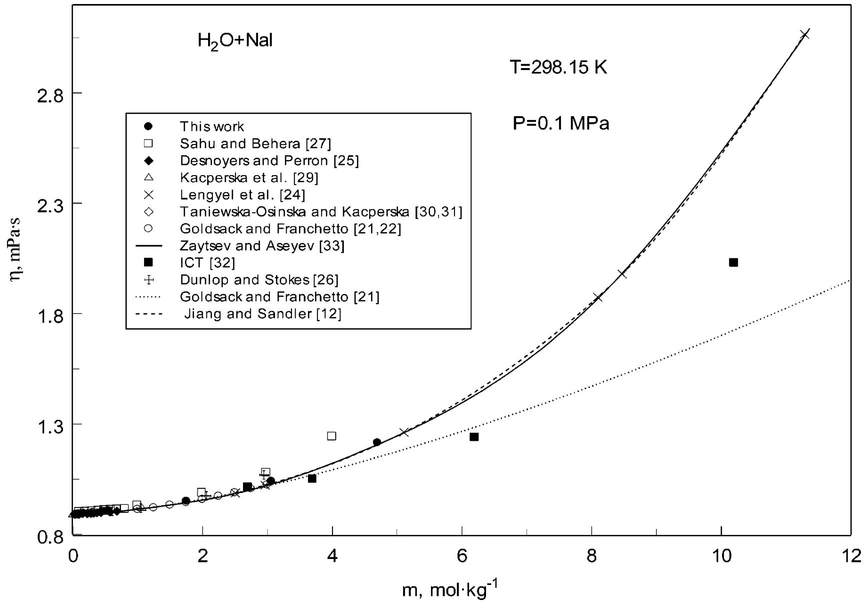

Figure 1. Measured values of viscosity η of NaI(aq) solutions as a function of molality m along the selected isotherm of 298.15 K and at atmospheric pressure together with values reported by other authors from the literature.

and can be determined by adding individual contributions of the solute constituents $B = z^{+}B^{-} + z^{-}B^{+}$, where $z^{\pm}$ and $B^{\pm}$ refer to the ionic valence and ionic (cationic and anionic) viscosity *B*-coefficient.[14,65−74] 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 $B^{\pm}$ at different temperatures are available in the literature (see, for example, refs 14 and 65−74). The behavior of the concentration dependence of the viscosity of solutions depends on the nature of the solute ions (Das,[72] Das and Das[73]). No theory has yet been developed for the *B*-coefficient. The *B*-coefficient is an adjustable parameter and is related to the size and the shape of the ions and ion−solvent and ion−ion interactions, respectively. Therefore, 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 aqueous electrolyte solutions is reported by various authors.[5−7,14,65,75−86] Low concentration viscosity measurements for most aqueous electrolyte solutions show good agreement between experiment and theory.[69,75−79,81] The values of the viscosity *A*-coefficient are always positive, while the *B*-coefficient can be either positive or negative depending on the nature of the solute and the solvent. 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 is associated with structure-breaking (disordering) ions.[14,83,84] The temperature coefficient of *B* is positive ($dB/dT > 0$), in most cases. The values of dB^{+}/dT for Mg^{2+} and Li^{+} are negative, for Na^{+} are almost zero, and for K^{+} are positive. The values of *B*-coefficients for I^{-} and Cl^{-} increase with temperature ($dB^{-}/dT > 0$) (Jenkins and Marcus[14]). The ions I^{-} interact weakly with the solvent molecules and act as structure-breakers. The viscosity *B*-coefficient for aqueous solutions shows strong temperature dependence, which can be attributed to ion−solvent interactions. However, measurements of the temperature dependence of the *A*- and *B*-coefficients have so far been limited to rather narrow ranges of temperature (at 298.15 K) with less than satisfactory accuracy. The viscosity *A*- and *B*-coefficients values of aqueous NaI solutions as a function of temperature in the temperature range from (298 to 575) K have been examined.

Desnoyers and Perron,[25] Kaminsky,[75−77] Jones and Talley,[78] Desnoyers et al.,[84] Feakins and Lawrence,[87] Robertson and Tyrrell,[88] and Chagnes et al.[89] added a quadratic term Dc^2 (extended Jones−Dole equation)

$$\eta_r = 1 + A\sqrt{c} + Bc + Dc^2 \tag{3}$$

to extend the Jones−Dole equation for more concentrated electrolyte solutions. The new Dc^2 term of eq 3 includes all solute−solvent and solute−solute structural interactions that were not accounted for by the $Ac^{1/2}$ and Bc terms at high concentrations such as:[5,25,84] 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 together with reported data for the relative viscosity, η_r, of NaI(aq) for various temperatures at low concentrations were used to calculate the values of the viscosity *A*-, *B*-, and *D*-coefficients in the extended Jones and Dole eq 3. The values of the viscosity *A*-, *B*-, and *D*-coefficients for NaI(aq) derived by least-squares analysis are summarized in Table 4 for two isobars (0.1 and 10) MPa. The derived values of the viscosity *A*- and *B*- coefficients are presented in Figure 4 as a function of temperature together with reported data. For the sake of comparison, *B*-coefficients reported by other authors and calculated from the ionic viscosity $B^{\pm}$-coefficient[14,80,67,68] have been included in Figure 4 (right). As one can see from Figure 4, generally the agreement between *A*- and *B*-coefficients derived from the present data and calculated with ionic $B^{\pm}$-coefficients data is good. Figure 4 (left) also demonstrates that the viscosity *A*-coefficient monotonically increases with temperature and the derivative (temperature coefficient) $dB/dT > 0$ is positive (structure-breaking ions).

The values of *A*- and *B*-coefficients can be estimated directly from experimental viscosity data as the slope of the dependence[25,73,90] $(\eta_r - 1)/c^{1/2}$ on $c^{1/2}$ (Jones−Dole plot, see Figure 5). Figure 5 illustrates the Jones−Dole plot for NaI(aq) solutions for the fixed temperature of 298.15 K. The intercepts and the slopes of the linear plots are the viscosity *A*- and *B*-coefficients, respectively. The derived values of the *A*- and *B*-coefficients

Table 4. Values of the Viscosity *A*-, *B*-, and *D*-Coefficients (Equation 4) of Aqueous NaI Solutions as a Function of Temperature

T/K	A/$\mathrm{dm^{3/2} \cdot mol^{-1/2}}$	B/$\mathrm{dm^3 \cdot mol^{-1}}$	D/$\mathrm{dm^6 \cdot mol^{-2}}$
298.15[a]	0.01085	0.0122	0.01266
323.15[a]	0.02180	0.0415	0.00307
333.15[a]	0.03145	0.0454	0.00374
343.15[a]	0.03570	0.0581	−0.00063
348.15[a]	0.03770	0.0627	−0.00121
353.15[a]	0.04110	0.0647	−0.00166
373.15[b]	0.05420	0.0742	−0.00479
398.15[b]	0.06765	0.0799	−0.00871
423.15[b]	0.07134	0.0901	−0.00620
448.15[b]	0.08513	0.0941	−0.00774
473.15[b]	0.09632	0.0966	−0.00833
498.15[b]	0.12050	0.0992	−0.01100
523.15[b]	0.13261	0.1011	−0.04720
548.15[b]	0.17510	0.1021	−0.09245
573.15[b]	0.17960	0.1051	−0.13777

[a] At a pressure of $P = 0.1$ MPa. [b] At a pressure of $P = 10$ MPa

for 298.15 K are 0.00279 $\mathrm{dm^{3/2} \cdot mol^{-1/2}}$ and 0.0281 $\mathrm{dm^3 \cdot mol^{-1}}$, respectively. The value of the *B*-coefficient at 298.15 K predicted by ionic $B^{\pm}$-coefficient data (Jankins and Marcus[14]) $B = 0.012$ $\mathrm{dm^3 \cdot mol^{-1}}$ is almost two times lower than the present value, while the values reported by Palepu et al.[91] (0.0317 $\mathrm{dm^3 \cdot mol^{-1}}$), Robertson and Stokes[80] (0.0178 $\mathrm{dm^3 \cdot mol^{-1}}$), and Bhattacharyya[67,68] (0.0200 $\mathrm{dm^3 \cdot mol^{-1}}$) are in satisfactory agreement with the present results. The agreement between the values of the *A*- and *B*-coefficients reported by Kacperska et al.[29] at 296.15 K $A = 0.0052$ and $B = 0.023$ $\mathrm{dm^3 \cdot mol^{-1}}$ (extrapolated values) and the present result is still acceptable. Unfortunately, there are no reliable experimental viscosity data for NaI(aq) at dilute concentrations for the accurate determination of the intercepts and slopes of the Jones−Dole plot at high temperatures. Therefore, the estimated uncertainty in derived values of the *A*- and *B*-coefficients is about 5−6 %. The values of the viscosity *A*- and *B*-coefficients calculated also as limiting slope and intercepts are shown in Figure 5 together with the data reported by other authors.

Lencka et al.[11] developed a comprehensive model for calculating the viscosity of aqueous electrolyte solutions ranging from dilute to very concentrated. The model includes a long-range electrostatic interactions term, η_r^{LR} (Onsager−Fuss theory), contributions of individual ions, η_r^{S} (using Jones−Dole *B*-coefficients), and a contribution of specific interactions between ions or neutral species, η_r^{S-S} (for the concentrated solutions, function of the ionic strength)

$$\frac{\eta}{\eta_0} = 1 + \eta_r^{LR} + \eta_r^{S} + \eta_r^{S-S} \tag{4}$$

where

$$\eta_r^{LR} = a\frac{1}{\eta_0}\left(\frac{2I}{\epsilon T}\right)^{1/2}\left[\left(\sum_i^{N_I}\frac{\mu_i z_i}{\lambda_i}\right) - 4r\sum_{n=0}^{\infty} c_n s^n\right] \tag{5}$$

$$\eta_r^{S} = \sum_i c_i B_i \tag{6}$$

$$\eta_r^{S-S} = \sum_i \sum_j f_i f_j D_{ij} I^2 \tag{7}$$

where $a = 0.36454$; η_0 and ϵ are the viscosity and dielectric constant of pure water (Kestin et al.,[57] Fernandez et al.[64]), respectively;$\mu_i = c_i z_i^2/\sum_j^{N_I} c_j z_j^2$; λ_i is the equivalent conductance of ion i; $r_i = 1 - z_i\lambda_i/(z/\lambda)$ ($i = 1, N_I$); and B_i are the molar concentration and *B*-coefficient of the *i*th species, respectively; f_i and f_j are fractions if the *i*th and *j*th species; D_{ij} is the interaction parameter between i and j. For concentrated solutions, the parameter D_{ij} depends on the ionic strength and can be expressed as

$$D_{ij} = d_1 + d_2 I + d_3 \exp(0.08 I^{3/2}) \tag{8}$$

where d_i are the empirical parameters. The model reproduces the viscosity of aqueous systems ranging from dilute to concentrated solutions (30 *m*) at temperatures up to 573 K. A technique for predicting the temperature dependence of the viscosity *B*-coefficients has been developed using the concept of structure-breaking and structure-making ions[11]

$$B = B_E + B_S \exp[-K(T - 273.15)] \tag{9}$$

where the value of $K = 0.026$ is a constant, $B_E = 0.0945$ $\mathrm{L \cdot mol^{-1}}$ is the Einstein contribution (hydrodynamic, intrinsic term), and $B_S = -0.1866$ $\mathrm{L \cdot mol^{-1}}$ was calculated using the present values of *B*-coefficients for NaI(aq). Comparisons of values of the viscosity *B*-coefficients calculated from eq 9 with the values reported in the literature are given in Figure 4.

The Hubbard−Onsager theory[92,93] of dielectric friction was applied by Ibuki and Nakahara[94] to the viscosity *B*-coefficients. The resulting expression for the viscosity *B*-coefficient is

$$B = \frac{4\pi N_A}{3000} r_{HO}^{\ 3} f\left(\frac{r}{r_{HO}}\right) \tag{10}$$

where r_{HO} is the characteristic radius defined as (Hubbard and Onsager[92])

$$r_{HO} = [(e^2\tau/16\pi\eta_S)(\epsilon_0 - \epsilon_\infty)/\epsilon_0^{\ 2}]^{1/4} \tag{11}$$

where e is the unit charge, τ is the dielectric relaxation time, ϵ_0 is the static permittivity (i.e., that at low frequency), ϵ_∞ is the high-frequency permittivity, and function $f(r/r_{HO})$ had to be calculated numerically from a solution of the differential equation of the shear viscosity.[92] For aqueous solutions, $r_{HO} = 0.15$ nm. The values of the $B^{\pm}$-coefficients for $\mathrm{Na^+}$ and $\mathrm{I^-}$ at 298.15 K calculated from this theory are about 0.060 and 0.018 $\mathrm{dm^3 \cdot mol^{-1}}$, respectively. Therefore, the predictive value of the *B*-coefficient for NaI(aq) at 298.15 K is about 0.078 $\mathrm{dm^3 \cdot mol^{-1}}$ which is considerably higher than the present and published experimental data. This theory is not successful in predicting the temperature dependence of the *B*-coefficient. This means that the dielectric and viscous frictions are minor factors in the temperature effect on the *B*-coefficient. Therefore, the structure and molecular nature of the solvent are important factors which define the temperature dependence of the *B*-coefficient.

The Einstein theory[95] is a well-known continuum theory for the *B*-coefficient. Einstein[95] calculated the increase of the viscosity of solution due to the perturbation of the original field by the presence of the solute in the solvent continuum. Einstein[95] calculated the increase in the viscosity from the linearization of the Navier−Stokes equation as

$$\eta_r = 1 + k\phi \tag{12}$$

where $\phi = (4/3)\pi R^3 N_A c$ is the volume fraction of the solute molecules, R is the effective solute ion radius, $k = 2.5$ is a constant determined by the hydrodynamic boundary condition

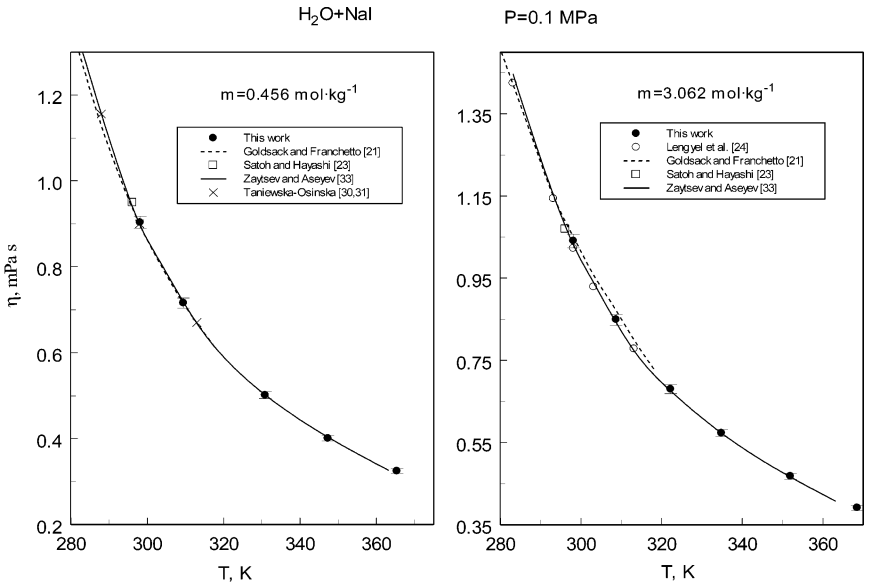

Figure 2. Measured values of viscosity η of NaI(aq) solutions as a function of temperature T along the two selected concentrations of (0.456 and 3.062) mol·kg^{-1} and at atmospheric pressure together with values reported by other authors from the literature.

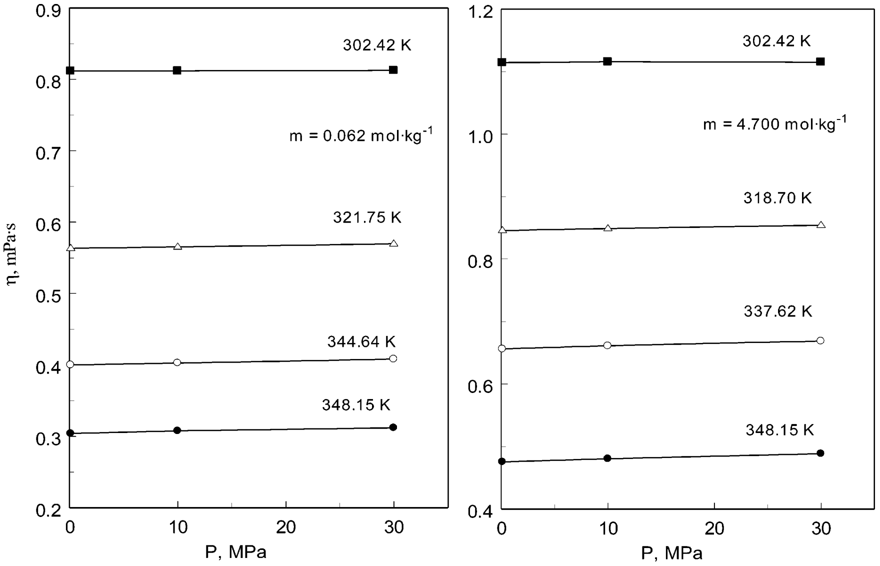

Figure 3. Measured values of viscosity η of NaI(aq) solutions as a function of pressure P at various temperatures at two selected concentrations (0.062 and 4.700) mol·kg^{-1}. Solid lines are predicted from eq 20.

at the solute surface. Thomas[96] has extended the limiting Einstein relation (eq 12) for the hydrodynamic effect to high concentrations by showing that for suspensions the relative viscosity is given by the relation

$$\eta_r = 1 + 2.5\phi + 10.05f^2 = 1 + 2.5V_kc + 10.05V_k^2c^2 \tag{13}$$

where $\phi = V_kc$ and V_k is the hydrodynamic molar volume or effective rigid molar volume of the salt. As was shown by Breslau and Miller,[97] this relation can be used to represent the concentration dependence of the relative viscosity for concentrated electrolyte solutions if V_k is taken as an adjustable parameter. We determined the values of V_k as a function of temperature for NaI(aq) in eq 13. The derived values of V_k are given in Table 5 and shown in Figure 6 (right). As one can see from this table, the value of $V_k = 0.0149$ L·mol^{-1} at 298.15 K is in satisfactory agreement with the value 0.0171 L·mol^{-1} by Breslau and Miller[97] but is much higher than the value of 0.008 L·mol^{-1} reported by Sahu and Behera[27] for NaI(aq). Therefore, eq 13 can be used to estimate the values of the hydrodynamic volume V_k from experimental relative viscosity data. Moulik and Rakshit[98] also used eq 13 to correlate the concentration dependence of the viscosity of 72 different electrolyte solutions at high concentrations. A least-squares fit of the correlation between viscosity B-coefficients and the hydrostatic volumes V_k for NaI(aq) results in the following correlation $B = 2.8502V_k - 0.0286$. Relating Einstein's eq 12 for the relative viscosity to the Jones−Dole eq 3, the B-coefficient can then be related to the molar volume V_k by $B = 2.5V_k$. Breslau and Miller[97] proposed the following equation for 1:1 electrolytes $B = 2.9V_k - 0.018$, which is very close to the present result. Isono and Tamamushi[99] also found a linear relation between the viscosity B-coefficient of the electrolyte solution and the molal volume V_k of the hydrated salt, $B = aV_k - b$, where the values of parameter a vary within (2.6 to 2.8) depending on temperature. As one can see, the values of a are very close to the Einstein value of 2.5. Our result for $a = 2.85$ is in good agreement with the values reported by Isono and Tamamushi[99] and close to the Einstein value of 2.5 for the ideal system. As discussed by Desnoyers and Perron,[25] V_k should be given by the partial molar

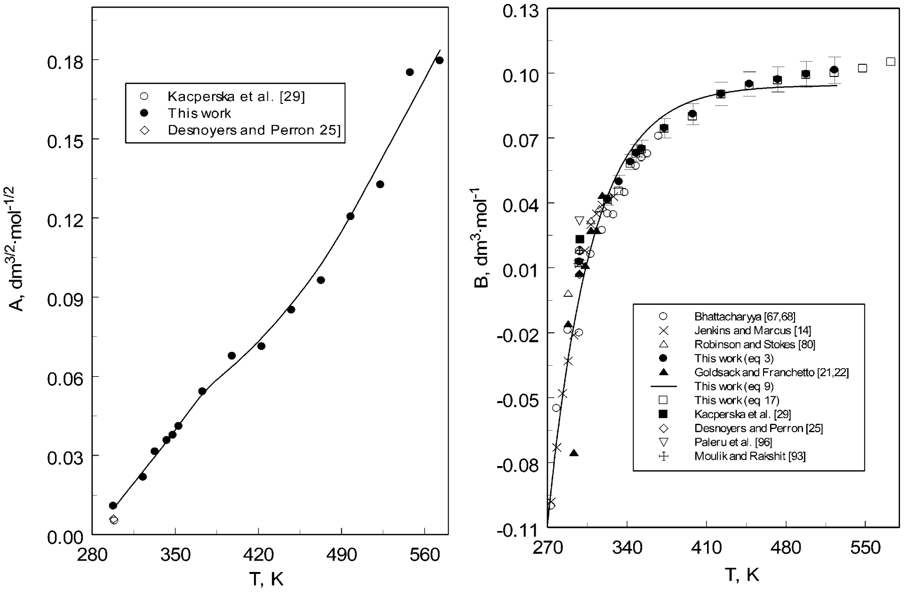

Figure 4. Experimental viscosity *A*- and *B*-coefficients of the NaI(aq) solutions as a function of temperature together with values reported by other authors from the literature.

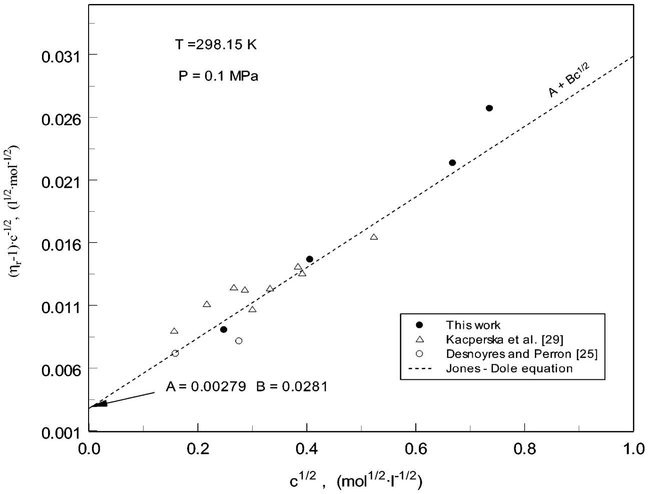

Figure 5. Jones−Dole plot, $(\eta_r - 1)/c^{1/2}$ vs $c^{1/2}$ for NaI(aq) solutions for the selected isotherm of 298.15 K at atmospheric pressure.

volume of the solute $\bar{V}$, although other authors (for example, Skinner and Fuoss[100]) considered V_k as the apparent molar volume. Therefore, for such an ideal system, the viscosities are predictable from densities and vice versa. Sahu and Behera[27] also represented the experimental relative viscosity of electrolyte solutions as

$$\eta_r = 1 + 2.5\bar{V}c + k_1\bar{V}^2c^2 + k_2\bar{V}^3c^3 + k_3\bar{V}^4c,^4 \qquad (14)$$

where $\bar{V}$ is the molar volume of electrolyte in solution ($dm^3 \cdot mol^{-1}$) and c is the concentration in $mol \cdot dm^{-3}$. The values of correlating coefficients k_i ($i = 1,3$) for electrolyte solutions were derived from experimental viscosity data at 25 °C (ref 27).

Table 5. Values of Effective Rigid Molar Volume (V_k, Equation 13) of NaI From Viscosity Data as a Function of Temperature at Atmospheric Pressure

T K	V_k $L \cdot mol^{-1}$
298.15[a]	0.01488
323.15[a]	0.02301
333.15[a]	0.02714
343.15[a]	0.03019
348.15[a]	0.03182
353.15[a]	0.03308
373.15[b]	0.03782
398.15[b]	0.04143
423.15[b]	0.04629
448.15[b]	0.07617
473.15[b]	0.08273
498.15[b]	0.09607
523.15[b]	0.10496
548.15[b]	0.11480
573.15[b]	0.12410

[a] At a pressure of $P = 0.1$ MPa. [b] At a pressure of $P = 10$ MPa

A simple extension of the absolute rate theory of viscosity to include the contributions from all the particles in a concentrated electrolyte solution led to an equation which quantitatively accounted for the concentration dependence of the viscosity of electrolyte solutions in the 1−10 *m* range at fixed temperature (Goldsack and Franchetto[21,22])

$$\eta = \frac{\eta_0 \exp(xE)}{1 + xV} \qquad (15)$$

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

$$V = \left(\frac{V_c + V_a}{V_0}\right) - 2 \qquad (16)$$

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

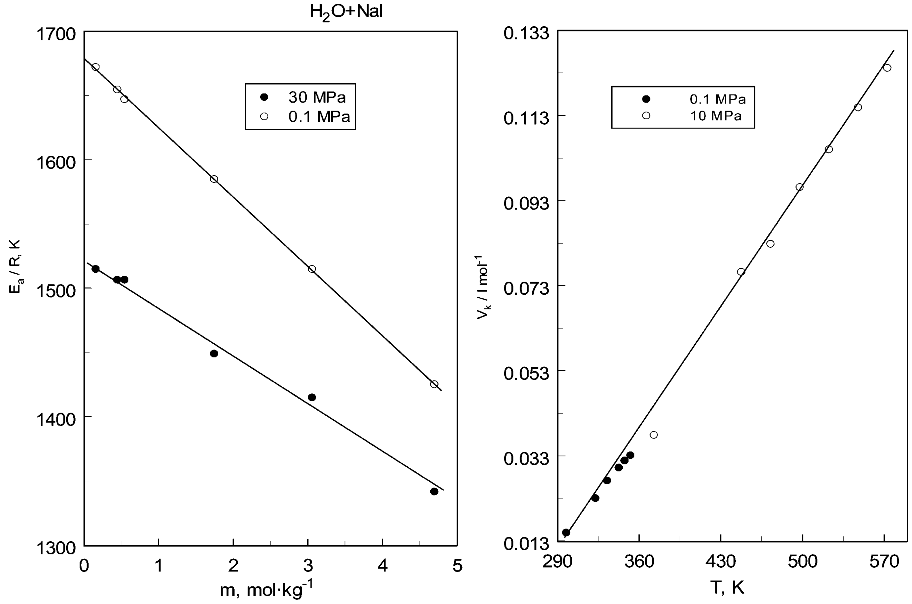

Figure 6. Values of flow activation energy, E_a/R (left), and effective rigid molar volume of NaI (hydrodynamic molar volume), V_k (right), calculated from the present viscosity data.

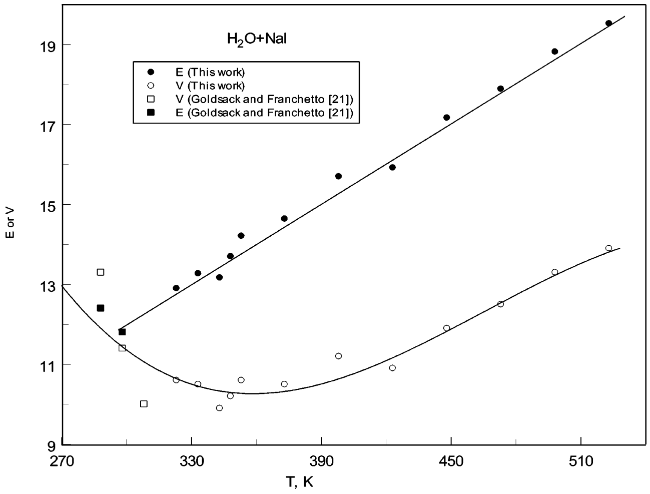

Figure 7. Values of parameters V and E (eq 15) for aqueous NaI solutions as a function of temperature. The solid curves are a guide for the eye.

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 molality of the salt. The individual ionic components of the V parameter are related to ionic solvation numbers, and the individual ionic free energy of activation components of the E are related to a surface free energy for formation of a hole in the liquid. Equation 15 was applied to the present experimental viscosity data for NaI(aq). The results are presented in Table 6 and shown in Figure 7. Analysis of the V parameters of the electrolytes leads to absolute ionic hydration numbers (Goldsack and Franchetto[21,22]). The temperature dependence of these hydration numbers reveals two types of ionic behavior: structure-making ions and structure-breaking ions.

The Jones−Dole viscosity B-coefficient is related to the parameters V and E by the relation

$$B = \frac{E - V}{55.51} \tag{17}$$

The values of the viscosity B-coefficients calculated with eq 17 using the values of V and E parameters for NaI(aq) are given in Table 6 and shown in Figure 4. The agreement between these data and the values derived by other authors at low temperatures is acceptable.

Table 6. Values of Parameters V and E (Equation 15) for Aqueous NaI Solutions as a Function of Temperature

T / K	E	V	B / dm^3·mol^{-1}
298.15	13.77	13.7	0.0128
323.15	12.90	10.6	0.0415
333.15	13.27	10.5	0.0498
343.15	13.17	9.90	0.0589
348.15	13.70	10.2	0.0631
353.15	14.21	10.6	0.0651
373.15	14.64	10.5	0.0746
398.15	15.70	11.2	0.0811
423.15	15.92	10.9	0.0904
448.15	17.17	11.9	0.0950
473.15	17.89	12.5	0.0971
498.15	18.82	13.3	0.0995
523.15	19.53	13.9	0.1014

Leyenndekkers[101] applied the TTG (Timmann−Tait−Gibson) model to viscosity to develop an equation covering zero to high (near or at saturation) concentrations. The derived equation covers changes in temperature and pressure and is simply extended to multicomponent solutions. The general principles of the TTG model have been outlined in the works of Leyenndekkers[101] and Leyenndekkers and Hunter.[102] The basic idea of the TTG model is that the effect of the ionic size, shape, and charge on water is equivalent to the effect of an applied pressure (the effective pressure P_e) which changes the volume of the water to the "apparent" volume of water in solution, ψ_w. The volume of the salt in the solution, ψ_S, is negligibly affected by pressure. According to the TTG model, the viscosity equation can be presented as[101,102]

$$\eta_S = \eta_{wis} \exp(a_1 m + a_2 m^2 + ...) \tag{18}$$

where η_S and η_{wis} represent the viscosities of the solution and the water in solution, respectively, and m is the concentration in molality. This equation can be interpreted to mean that at a

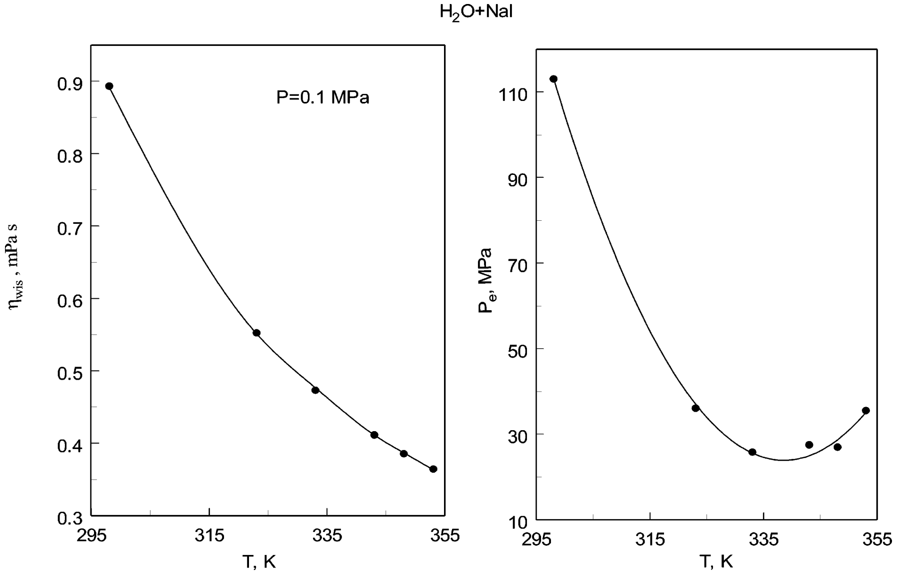

Figure 8. Values of viscosity of water in solution, η_{wis} (left), and effective pressure due to the salt, P_e (right), calculated with eqs 18 and 19, respectively, from the present viscosity data at atmospheric pressure: (—), smoothed curve.

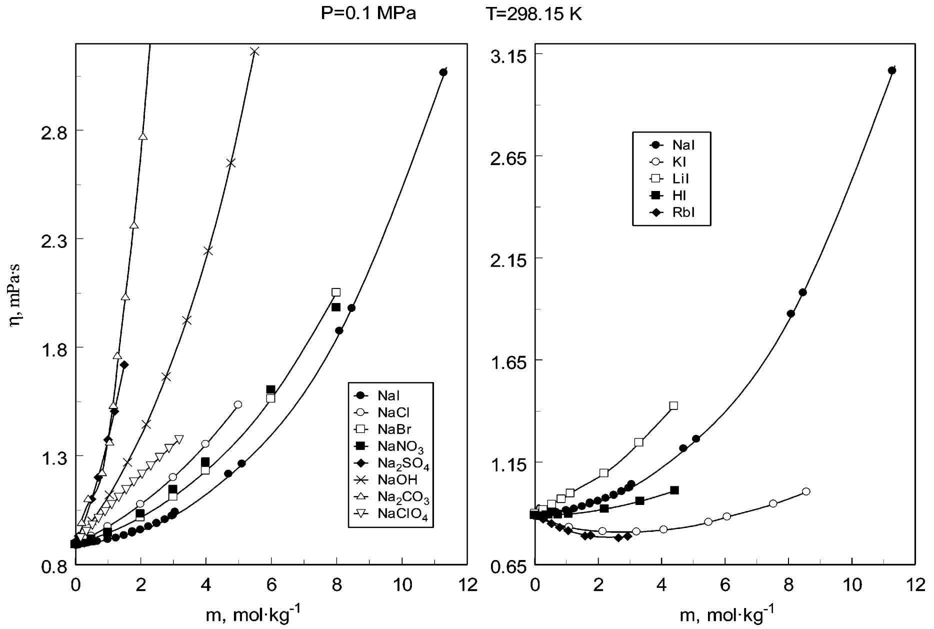

Figure 9. Viscosity of a series of aqueous solutions as a function of composition at a selected temperature of 298.15 K and a pressure of 0.1 MPa reported by various authors. Left: ●, NaI (this work); ○, NaCl (Isono[110,111]); □, NaBr (Isono[110,111]); ■, $NaNO_3$(Isono[110,111]); ◆, Na_2SO_4 (Isono[110,111]); ×, NaOH (Zaytsev and Aseyev[33]); △, Na_2CO_3 (Zaytsev and Aseyev[33]). Right: ●, NaI (this work); ○, KI (Lengyel et al.[24]); ○, LiI (Stokes and Mills[5]); ◆, RbI (Getman[112] and Desnoyers and Perron[25]); ■, HI (Zaytsev and Aseyev[33]).

given temperature the difference in the phenomenological activation volumes (Kell[103]) of the solution and the water in solution is a simple function of the concentration. It can be expected that the parameters $f(m)$, the TTG parameters, will be simple functions of the solute properties, in this case, the TTG volume. The value of η_{wis} is given by

$$\eta_{wis}(P) = \eta_w(P_e + P) \tag{19}$$

where η_w is the viscosity of pure water at pressure ($P + P_e$), P is the external pressure, and P_e is the effective pressure due to the salt. For each electrolyte, the value of a_1 is close to the corresponding Jones–Dole coefficient (viscosity B-coefficient). On the basis of the TTG model, the values of $f(m)$ should not be very sensitive to pressure. Thus, the viscosity of the solution at pressure P is given by

$$\frac{\eta_S(P)}{\eta_S(0.1)} = \frac{\eta_w(P_e + P)}{\eta_w(P_e + 0.1)} \tag{20}$$

Equation 18 was used to calculate the values η_{wis} for NaI(aq) from the present experimental viscosity data for the solution. This equation accurately represents the concentration dependence of the viscosity of aqueous solutions if η_{wis} is considered as an adjustable parameter. The derived values of η_{wis} are given in Table 7 and shown in Figure 8 (left). The values of the effective pressure P_e were calculated from eq 19 using derived values of η_{wis} (left side of eq 19) and the IAPWS[57] reference equation for the viscosity of pure water (right side of eq 19) by using an iteration procedure. The derived values of P_e are presented in Table 7 for various temperatures and shown in Figure 8 (right). Afzal et al.[104] also used this relation to describe their experimental viscosity data for some aqueous electrolyte solutions.

Table 7. Values of the Viscosity of Water in Solution (η_{wis}, Equation 18), the Parameters (a_1,a_2), and the Effective Pressure P_e of NaI From Viscosity Data as a Function of Temperature

T / K	η_{wis} / mPa·s	a_1 / kg·mol^{-1}	a_2 / kg^2·mol^{-2}	P_e / MPa
298.15[a]	0.8925	0.02036	0.00966	112.9
323.15[a]	0.5518	0.03932	0.00806	26.0
333.15[a]	0.4724	0.04997	0.00687	25.7
343.15[a]	0.4109	0.05308	0.00687	27.4
348.15[a]	0.3848	0.05559	0.00726	26.9
353.15[a]	0.3638	0.05038	0.00925	35.4
373.15[b]	0.2922	0.06024	0.00861	29.3
398.15[b]	0.2323	0.06083	0.01046	30.0
423.15[b]	0.1905	0.09153	0.00191	23.0
448.15[b]	0.1635	0.07193	0.01140	27.6
473.15[b]	0.1434	0.07014	0.01279	30.0
498.15[b]	0.1273	0.07576	0.01282	28.3
523.15[b]	0.1140	0.08401	0.01217	25.0

[a] At a pressure of $P = 0.1$ MPa. [b] At a pressure of $P = 10$ MPa

Figure 9 (right) demonstrated the concentration dependence of the viscosity of a series of aqueous solutions with the same anions (I^-) and various cations (K^+, Na^+, Li^+, Rb^+, and H^+). This figure demonstrates the effect of various cations on the values and concentration dependence of the viscosity of salt solutions (iodides). As one can see from Figure 9 (right), the NaI(aq) solutions observed intermediate values of viscosity among other aqueous (KI, LiI, HI, and RbI) solutions at the same thermodynamic (P,T,m) conditions. The viscosity of RbI(aq) and KI(aq) solutions at constant temperature and pressure decreases with concentration and passes through a minimum between (2 and 3) mol·kg^{-1} and then increases at higher concentrations. Figure 9 (left) also shows the concentration dependence of the viscosity of a series of aqueous solutions with the same cation (Na^+) and various anions (SO_4^{-2}, NO_3^-, Cl^-, Br^-, I^-, ClO_4^-, OH^-, and CO_3^{-2}). Figure 9 (left) shows the NaI(aq) observed lowest values of viscosity among other solutions (NaCl, NaBr, NaI, Na_2SO_4, $NaNO_3$, Na_2CO_3, $NaClO_4$, and NaOH). Figure 9 also demonstrates how the behavior of the concentration dependence of the viscosity of solutions depends on the nature of the solute ions. The different concentration behavior (see Figure 9) for several electrolyte solutions indicates the dependence of ionic interactions on the nature of the electrolytes.

Temperature Dependence of the Viscosity of NaI(aq) Solutions. The selected experimental viscosity data for NaI(aq) solutions as a function of temperature are shown in Figure 2 in the $\eta-T$ projection together with values reported by other authors at constant pressures and constant composition. The viscosity of solution considerably decreases with temperature. For example, at constant pressures (from 0.1 to 30 MPa) between temperatures (298 and 575) K, the viscosity of NaI(aq) solutions changes by (5 to 9) %. For pure water at the same temperature and pressure ranges, the viscosity changes by 11 %. In the concentration range $m > 1$ mol·kg^{-1}, the empirical equation of Arrhenius−Andrade is valid for the temperature dependence of the viscosity[5,105−108]

$$\eta = \eta_0 \exp\left(\frac{b}{T}\right) \tag{21}$$

where η_0 and $b = E_a/R$ (E_a is the flow activation energy) are a function of concentration. The values of these parameters for NaI(aq) solution calculated with the present measurements at pressures (0.1 and 30) MPa and for each measured concentration are given in Table 8 together with values for pure water. It follows from Table 8 that the values of the flow activation energy (E_a/R) decrease monotonically along the isobars (0.1 and 30) MPa (see Figure 6 left). Chagnes et al.[89,109] proposed a linear relation between the activation energy for the viscous flow E_a and the salt concentration as

Table 8. Values of the Arrhenius Parameters at Pressures of 0.1 and 30 MPa for NaI(aq) (Equation 21)

	$P = 0.1$ MPa		$P = 30$ MPa	
m / mol·kg^{-1}	$\ln \eta_0$	b / K	$\ln \eta_0$	b / K
0.000	−5.8842	1712.31	−5.7365	1667.18
0.062	−5.8492	1705.06	−5.6201	1632.53
0.166	−5.7400	1671.87	−5.2581	1514.76
0.456	−5.6693	1654.44	−5.2133	1506.25
0.552	−5.6385	1646.87	−5.2027	1506.36
1.754	−5.3871	1584.62	−4.9650	1448.88
3.062	−5.0639	1514.72	−4.7544	1414.84
4.700	−4.6048	1425.07	−4.3495	1341.56

$$E_a = E_a^0 - E_a^S m \tag{22}$$

where E_a^0 and E_a^S are the energy of activation for the pure solvent and the contribution of the salt to the activation energy for the transport process, respectively. Figure 6 (left) demonstrates the variation of E_a with concentration for two pressures (0.1 and 30) MPa. The derived values of E_a^0 and E_a^S are the following: 13.96 kJ·mol^{-1}·K^{-1} and 0.448 kJ·kg^{-1}·K^{-1} (at $P = 0.1$ MPa) and 12.66 kJ·mol^{-1}·K^{-1} and 0.316 kJ·kg^{-1}·K^{-1} (at $P = 30$ MPa). Equation 21 fails to represent the present data over the complete temperature range (at high temperatures $T > 373$ K). To accurately describe the experimental viscosity data at high temperatures (up to 575 K), one more term is required[105,106]

$$\eta = \eta_0 \exp\left(\frac{b}{T} + \frac{c}{T^2}\right) \tag{23}$$

Equation 21 represents the present experimental viscosity data within 0.95 % at temperatures up to 373 K. The Eyring's absolute rate theory (Glasstone et al.[108]) enables a detailed description of the temperature dependence of the viscosity of concentrated aqueous electrolyte solutions

$$\eta = \frac{hN}{V} \exp\left(\frac{\Delta G^+}{RT}\right)$$

or

$$\eta = A \exp\left(\frac{\Delta H^+}{RT}\right) \tag{24}$$

where ΔG^+ and ΔH^+ are the free enthalpy of activation and enthalpy of activation, 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 ΔH^+ can be calculated from the slope of the straight line by the $\ln \eta \approx 1/T$ function (see Figure 10). 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 of eq 15 for the viscosity of solutions (see Figure 7).

Pressure Dependence of the Viscosity of NaI(aq) Solutions. The viscosity is affected little (up to 8 %) at high temperatures (575 K) and low concentrations (0.062 mol·kg^{-1}) and up to 1.5−2.7 % at low temperatures (302 K) by pressure (with the pressure changing between 0.1 and 30 MPa) (see Figure 3).

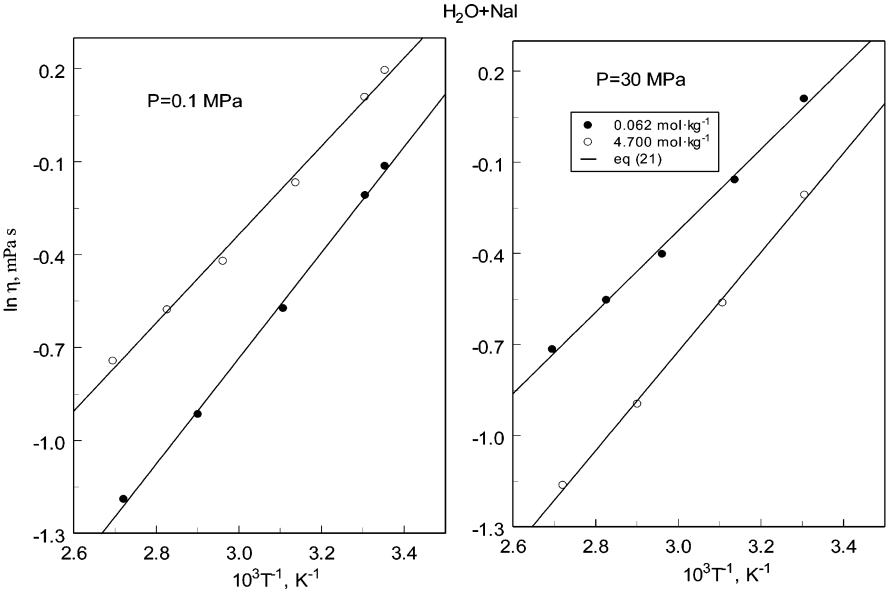

Figure 10. Measured values of ln η as a function of $1/T$ (Arrhenius−Andrade plot, left) at the two selected isobars (0.1 and 30) MPa for two constant concentrations (0.065 and 4.700) mol·kg^{-1} together with values calculated with eq 21.

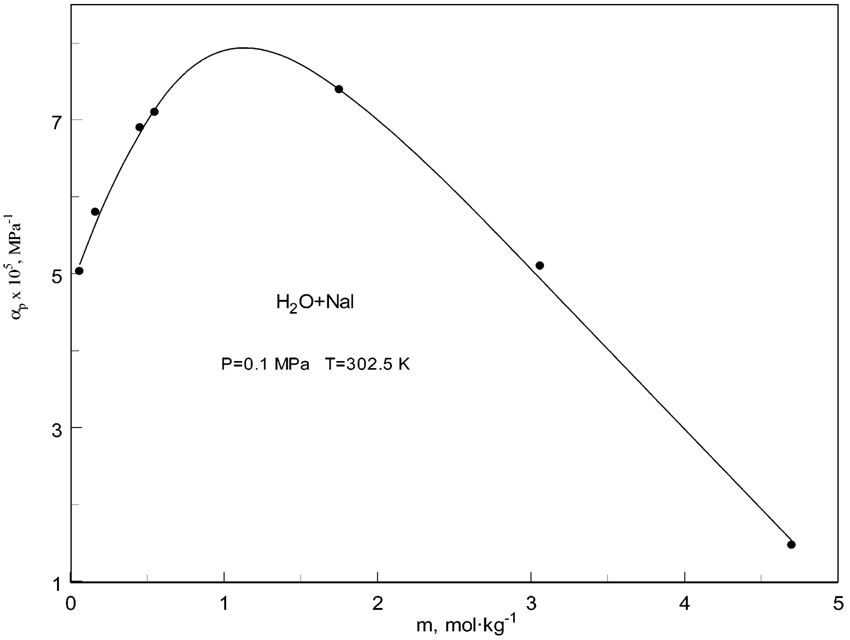

Figure 11. Pressure coefficients of viscosity, $\beta_P = \eta^{-1}(\partial\eta/\partial P)_{T,m}$, for NaI(aq) solutions at fixed pressure of 0.1 MPa and temperature 302.5 K as a function of concentration: (—), smoothed experimental curve.

Table 9. Values of the Pressure Coefficient for NaI(aq) at 0.1 MPa as a Function of Temperature for Selected Concentrations

$m = 0.062$ mol·kg^{-1}		$m = 4.700$ mol·kg^{-1}	
T	$(\partial \ln \eta/\partial P)_{T,m} \times 10^5$	T	$(\partial \ln \eta/\partial P)_{T,m} \times 10^5$
K	MPa^{-1}	K	MPa^{-1}
302.42	5.0320	302.50	1.4700
321.75	37.502	318.70	32.934
344.64	67.850	337.62	62.323
367.46	89.110	353.74	79.233
		370.98	91.382

The pressure dependence of the experimental viscosity of NaI(aq) in the range from (0.1 to 30) MPa is almost linear. The pressure dependence of the viscosity of aqueous solutions can be predicted by the TTG model (see eq 20). We applied eq 20 to NaI(aq) to calculate the viscosity at high pressures. The calculations were performed using the P_e from Table 7 and IAPWS[57] formulation for pure water. Figure 3 shows the comparison between measured and calculated values of the viscosity of NaI(aq) as a function of pressure along various isotherms and constant concentrations. As one can see, the agreement between predicted and measured values of the viscosity of NaI(aq) is excellent (deviations within 0.5 to 0.7 %). Figure 11 shows the concentration dependence of the experimental pressure coefficients of viscosity, $\alpha_P = \eta^{-1}(\partial\eta/\partial P)_{T,m}$, for NaI(aq) solutions at a fixed pressure of 0.1 MPa and temperature 302.5 K. As one can see from Figure 11, α_P goes through the maximum near 1.0 mol·kg^{-1}.

Conclusions

The present viscosity data for NaI(aq) agree with the most reported values within (0.2-0.9) % at 298 K and (1.2−1.4) % at temperatures of 313 K. The viscosity of NaI(aq) solutions increases monotonically very slightly with the electrolyte concentration without a minimum at low concentrations (up to

1.5 mol·kg^{-1}), then increases rapidly at high concentrations. The derived viscosity *A*-coefficients for NaI(aq) solutions monotonically increasing with temperature and the temperature coefficient d*B*/d*T* > 0 is positive (structure-breaking ions). The value of the *B*-coefficient at 298 K predicted by ionic $B^{\pm}$-coefficient data is in satisfactory agreement with the present measurements. The extended Einstein relation can be used to accurately estimate the values of the hydrodynamic volume V_k from the experimental relative viscosity data. The linear relation $B = aV_k - b$ (where $a = 2.8502$ and $b = 0.0286$) between the viscosity *B*-coefficients and the hydrodynamic volumes V_k was found. The value of *a* is close to the Einstein value of 2.5 and the value reported by other authors ($a = 2.9$). The TTG model can be applied to accurately predict the viscosity of NaI(aq) solutions at high temperatures and high pressures. It was found that the absolute rate theory of viscosity for aqueous electrolyte solutions with temperature depending parameters *V* and *E* is suitable to accurately represent the experimental viscosity data in the wide temperature (up to 575 K) and concentration (1−10) *m* regions. From the measured viscosity data, it can be concluded that the pressure coefficient of viscosity α_P increases with concentration, passes through a maximum near 1.0 mol·kg^{-1}, and then decreases at higher concentrations.

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