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Reviews

Model for Calculating the Viscosity of Aqueous Solutions

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A new model for calculating the viscosity of aqueous solutions has been developed. Parameters for 74 solutes were established based on a critical review of the literature for solutions of one solute in water, with over 9000 points included. The average difference between the calculated and experimental viscosities is less than 0.1 %, and the standard deviation of this difference is 3.7 % of the average experimental viscosity. The model was validated by estimating published viscosity for systems of more than one solute in water. The average difference between experimental and calculated values for 1700 points is −2.7 %, and the standard deviation of this difference is 16 % of the average experimental viscosity. The median standard deviation of the difference between experimental and calculated values is 3.5 % of the experimental viscosity. The solutes studied are $(NH_4)_2SO_4$, $AlCl_3$, $BaCl_2$, $Ca(NO_3)_2$, $CaCl_2$, $Cd(NO_3)_2$, $CdCl_2$, $CdSO_4$, $CoCl_2$, $CoSO_4$, $Cr_2(SO_4)_3$, $CrCl_3$, $Cu(NO_3)_2$, $CuCl_2$, $CuSO_4$, $Fe_2(SO_4)_3$, $FeCl_2$, $FeSO_4$, H_2O_2, H_2SO_4, H_3PO_4, HCH_3CO_2 (acetic acid), $HCHO_2$ (formic acid), HCl, HCN, HNO_3, K_2CO_3, $K_2Cr_2O_7$, K_2HPO_4, K_2SO_4, K_3PO_4, KBr, KCH_3CO_2, $KCHO_2$, KCl, KH_2PO_4, KI, KNO_3, KOH, Li_2SO_4, LiCl, $LiNO_3$, LiOH, $Mg(NO_3)_2$, $MgCl_2$, $MgSO_4$, $MnCl_2$, $MnSO_4$, Na_2CO_3, Na_2HPO_4, $Na_2S_2O_3$, Na_2SO_3, Na_2SO_4, Na_3PO_4, NaBr, $NaCH_3CO_2$, NaCl, $NaClO_3$, NaF, NaH_2PO_4, NaI, $NaNO_3$, NaOH, NH_3, NH_4Cl, NH_4NO_3, $NiCl_2$, $NiSO_4$, $Pb(NO_3)_2$, $Sr(NO_3)_2$, $SrCl_2$, sucrose, $ZnCl_2$, and $ZnSO_4$. Density data are also presented for these solutes and for $NaHCO_3$.

Introduction

Physical properties of aqueous solutions have countless practical applications. There is however no easily available source of critically evaluated data. In a previous review,[160] we presented data on the density of 59 electrolytes in water. In this review, we present data on the viscosity of 74 solutes in water. This review includes non-electrolyte solutes such as sucrose and hydrogen peroxide. Density data are also presented in the Supporting Information for all solutes covered in this review.

In the vast majority of practical applications, we find ourselves in a situation where we have to deal with a mixture of solutes. There is no widely accepted model that allows the calculation of the viscosity of such a mixture over a range of temperatures and concentrations. This review proposes such a model. Data on solutions of many solutes in water are used to validate its accuracy.

As in our previous review, all data are converted in consistent units, mass fraction for concentration, mPa•s for viscosity, and °C for temperature. In many instances, however, the original data use units that require the knowledge of the solution density for conversion. Such is the case when the concentration is expressed in molarity or when the viscosity was measured as kinematic viscosity. If the original data do not include this density, the correlation developed in our previous review is used. If the original data do include the density, then this value is used in the conversion.

Review of Available Data

The data used in this review come from a number of sources. As before, Lobo's *Handbook of Electrolyte Solutions*[165] was extensively used for data between the late 1800s and 1985. This Journal was also systematically reviewed from its beginning to today, as were recent issues of the *Canadian Journal of Chemistry* and the *Journal of Solution Chemistry*. Searches in Chemical Abstracts were also done for solutes that are not well-covered in these sources.

Review of Existing Viscosity Mixing Rules and Models

A large number of viscosity models were proposed over the years. In a 1977 review, Irving[122] presented 25 mixing equations, while in a 1995 review, Monnery et al.[181] presented over 20 different models using 230 different equations. It is not the scope of this review to go over all published equations and models, but some background is useful.

In this review, we distinguish between the *mixing rule* and the *viscosity model*. The mixing rule enables the calculation of the solution viscosity knowing the viscosity of its constituents. Conversely, the mixing rule may be used to calculate a constituent viscosity if the mixture viscosity and all other constituent viscosities are known. The viscosity model is used to predict the constituents' viscosity given their concentration and the solution temperature.

The general form of the mixing rule is

$$f(\eta_m) = \sum y_i f(\eta_i) \tag{1}$$

where y_i is the mole, mass, or volume fraction of component i; η_m is the solution viscosity ("m" here standing for "mixture"); η_i is the viscosity of the component i; and $f(\eta_m)$ is usually either η_m, $1/\eta_m$, or ln η_m.

It has been established[191,122] that the mixing rules that are additive, such as

$$\eta_m = \sum y_i \eta_i \tag{2}$$

or

$$1/\eta_m = \sum y_i/\eta_i \tag{3}$$

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do not predict correctly the viscosity of mixtures, especially of aqueous solutions. Logarithmic mixing rules, such as the Arrhenius rule,[15] are more successful:

$$\ln \eta_{\mathrm{m}} = \phi_1 \ln \eta_1 + \phi_2 \ln \eta_2 \tag{4}$$

where ϕ_i is the volume fraction of component i.

The Arrhenius mixing rule, however, does not allow for local minima or maxima in mixture viscosities. While this may be acceptable for ideal mixtures, this is definitively not satisfactory for mixtures of dissimilar components. This has long been recognized, and a number of mixing rules have been published to improve on the Arrhenius mixing rule to enable the correct representation of these minima and maxima.

Irving reviewed 54 mixing rules and extensively tested 25 of them. Irving defined a "goodness factor" equal to 20 if the rms error was over 10 % and 100 if it was below 2 %. He tested the mixing rules against 69 combinations of temperature and aqueous mixtures containing acetaldehyde, acetic acid, acetone, acetonitrile, butyric acid, dioxane, ethanol, ethyleneglycol, formamide, formic acid, glycerol, methanol, propanol, isopropanol, pyridene, and triethylamine.

Perhaps the best mixing rule tested by Irving for aqueous systems is the Grunberg−Nissan mixing rule[103] in which the mixture viscosity is expressed as follows:

$$\ln \eta_{\mathrm{m}} = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12} \tag{5}$$

where x_i is the mole fraction of component i and G_{ij} is an empirical adjustment parameter. In Irving's test, this mixing rule was found to have a goodness of fit of 29. However, modifying eq 5 to use the mass fraction increased the goodness of fit to 42, and using volume fraction increased this goodness of fit to 46. (A goodness of fit of 46 corresponds to a rms error from 6 % to 7 %.)

In general, Irving found that using mole fraction instead of volume or mass does not improve significantly on the accuracy of the calculated mixture viscosity. In fact, the opposite is true, and mass or volume fractions generally give better results. This was true of most mixing rules and systems studied, including nonaqueous systems.

The Grunberg−Nissan mixing rule however suffers from three problems when trying to calculate the viscosity of aqueous mixtures of arbitrary composition and number of components. The "pure component" viscosity of electrolytes cannot be determined directly. Infinite dilution viscosity could be used instead, but this is experimentally difficult to establish. All of Irving[122] aqueous systems were mixtures of miscible organic liquids and water. Second, calculating the viscosity of mixtures containing an arbitrary number of components is cumbersome, as the number of adjustment parameters G_{ij} increases as the square of the number of components (neglecting interaction parameters for three components or more). Finally, predicting the value of G_{ij} for mixtures of arbitrary composition and temperature is challenging. Some viscosity models that predict the value of G_{ij} are so complex as to be practically useless.

A modified form of the Arrhenius mixing rule was found to be more satisfactory for use in aqueous solutions. This mixing rule assumes that the viscosity of water η_{w} is a function of temperature only:

$$\eta_{\mathrm{w}} = f(t) \tag{6}$$

while the solutes viscosity η_i is variable and a function of both temperature and concentration:

$$\eta_i = f(t, w) \tag{7}$$

The mixing rule used herein is simply

$$\ln \eta_{\mathrm{m}} = w_{\mathrm{w}} \ln \eta_{\mathrm{w}} + \sum w_i \ln \eta_i \tag{8}$$

or, alternatively

$$\eta_{\mathrm{m}} = \eta_{\mathrm{w}}{}^{w_{\mathrm{w}}} \prod \eta_i^{w_i} \tag{9}$$

where w_{w} is the mass fraction of water and w_i is the mass fraction of solute i, being understood that for a system with n solutes, $w_{\mathrm{w}} + \Sigma_{i=1}^{n} w_i = 1$.

Therefore, for a solution of one solute in water, knowing the solution viscosity η_{m} and the solute mass fraction w_i, the solute viscosity η_i is simply

$$\eta_i = \left(\frac{\eta_{\mathrm{m}}}{\eta_{\underline{\mathrm{W}}}{}^{w_{\mathrm{w}}}} \right)^{1/w_i} \tag{10}$$

Equation 10 is used to calculate a solute viscosity using only viscosity data for solutions of this solute in water. Once the viscosities of a sufficient number of solutes have been calculated in this way, the viscosity of a solution of an arbitrary number of solutes can then be determined using eq 9. The solution viscosity calculated by eq 9 tends to the viscosity of water as the solutes concentration tends toward 0, and eq 9 is mathematically smooth and lends itself to both interpolation and extrapolation.

Model for the Viscosity of Pure Water. In order to evaluate η_i, a model is required to calculate the viscosity of water as a function of temperature. The International Association for the Properties of Water and Steam has published equations to this effect.[121] However, these equations are cumbersome and difficult to program in Excel, the software used to assemble and convert the data. The following correlation is used instead:

$$(\eta_{\mathrm{w}}/\mathrm{mPa{\cdot}s}) = \frac{t/{}^{\circ}\mathrm{C} + 246}{(0.05594t/{}^{\circ}\mathrm{C} + 5.2842)t/{}^{\circ}\mathrm{C} + 137.37} \tag{11}$$

Viscosities from *Recommended Reference Materials for the Realization of Physicochemical Properties*[171] between 0 °C and 100 °C were used to establish this correlation. Values from IAPWS between 110 °C and 150 °C were used to validate the goodness of fit of eq 11 and its ability to extrapolate beyond the range of the fitted data. Data from ref 171 are at atmospheric pressure; data from IAPWS are at saturation temperature. As can be seen in Table 1, the fit is excellent with a difference

Table 1. Comparison between Recommended and Fitted Values for the Viscosity of Water

t/°C	reference	recommended value of η_{w}/mPa·s	difference between recommended and calculated value/mPa·s
0	Marsh[171]	1.7910	0.0002
10	Marsh	1.3070	−0.0004
20	Marsh	1.0020	−0.0002
25	Marsh	0.8902	0.0000
30	Marsh	0.7975	0.0004
40	Marsh	0.6530	0.0004
50	Marsh	0.5469	0.0002
60	Marsh	0.4665	−0.0001
70	Marsh	0.4042	−0.0002
80	Marsh	0.3551	0.0000
90	Marsh	0.3150	−0.0002
100	Marsh	0.2821	−0.0003
110	IAPWS[121]	0.2547	−0.0004
120	IAPWS	0.2321	0.0000
130	IAPWS	0.2129	0.0004
140	IAPWS	0.1965	0.0009
150	IAPWS	0.1825	0.0016

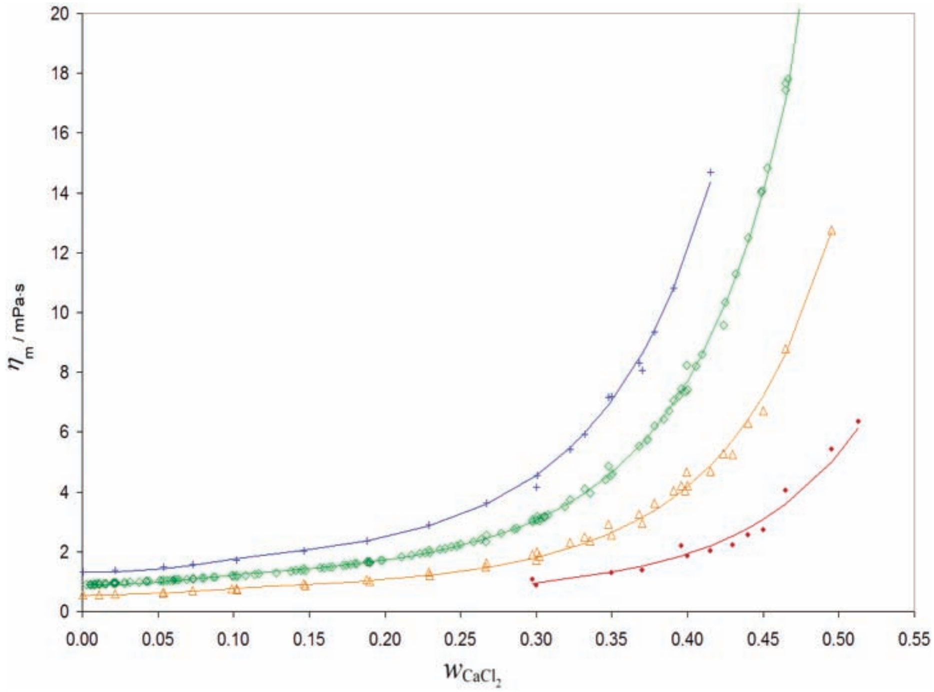

Figure 1. Viscosity of calcium chloride aqueous solutions at various temperatures: blue +, experimental at 10 °C; blue line, calculated at 10 °C; green ◊, experimental at 25 °C; green line, calculated at 25 °C; orange △, experimental at 50 °C; orange line, calculated at 50 °C; red ○, experimental at 90 °C; red line, calculated at 90 °C. Note the close fit between experimental and calculated viscosities at low concentration.

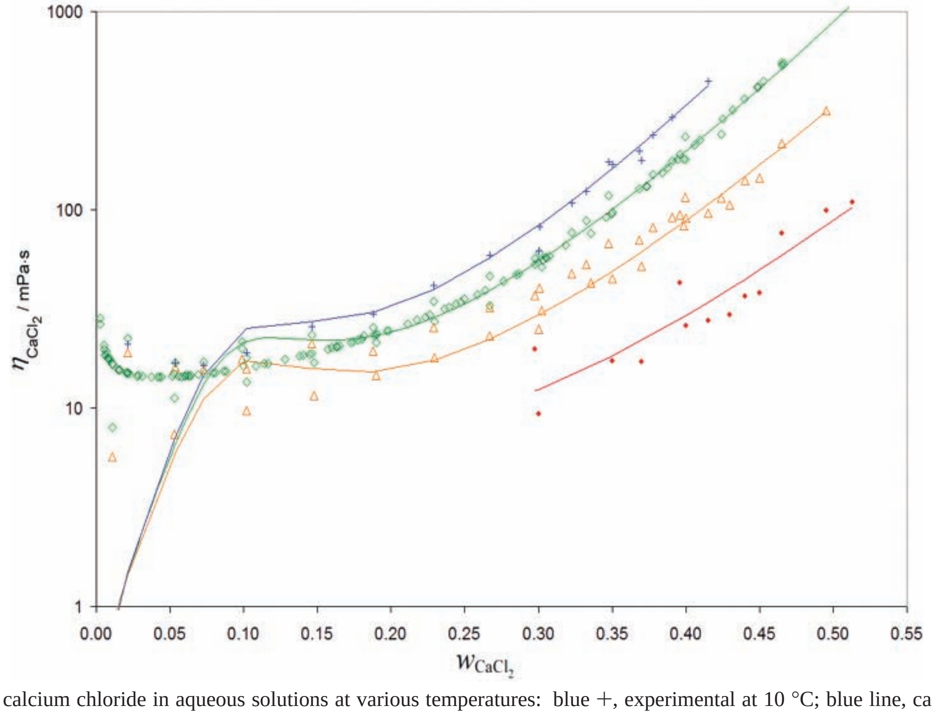

Figure 2. Viscosity of calcium chloride in aqueous solutions at various temperatures: blue +, experimental at 10 °C; blue line, calculated at 10 °C; green ◊, experimental at 25 °C; green line, calculated at 25 °C; orange △, experimental at 50 °C; orange line, calculated at 50 °C; red ○, experimental at 90 °C; red line, calculated at 90 °C. Note the wide variation between experimental data points for the same temperature. Also compare the underestimation of the calcium chloride viscosity at low concentration with the fit of the solution viscosity in Figure 1.

that does not exceed 0.1 % of the recommended value and is often 0.01 % or lower. Even when extrapolated to 150 °C the difference is still less than 1 %.

At high temperature there is a systematic under evaluation of the viscosity that is due to the effect of pressure. This effect is neglected in this review, as it is essentially non measurable (less than 1 %) at pressures below 0.5 MPa. As the pressure increases, however, its effect becomes noticeable. To generalize the use of eq 9 at high pressure it would be necessary to account for this factor in the evaluation of the viscosity of water.

Model for the Solute's Viscosity. Finding a model to correctly represent the viscosity of aqueous solutions is a challenge. Consider the case of $CaCl_2$ (see Figure 1). Six data sets are available for this solute (Afzal,[6] Bogatykh,[25] Isono,[124] Wahab,[288] Wimby,[292] Zang[298]). The viscosity of the solution increases slowly up to a mass fraction of about 0.2 and then starts increasing sharply. Figure 2 represents the viscosity of $CaCl_2$, calculated using the mixing rule from eq 9 and the viscosity of pure water from eq 11. A log scale is used to put emphasis on the viscosity at low concentration.

Table 2. Results for Solutions of One Solute in Water[a]

name	$(NH_4)_2SO_4$	$AlCl_3$	$BaCl_2$	$Ca(NO_3)_2$	$CaCl_2$	$Cd(NO_3)_2$	$CdCl_2$	$CdSO_4$	$CoCl_2$	$CoSO_4$	$Cr_2(SO_4)_3$	$CrCl_3$
v_1	11.567	−8335.5	2.9763	218.21	32.028	9.2559	6.5301	34.268	16.08	26.343	12.673	−120.2
v_2	2.5709	4823.7	1.104	5.4442	0.78792	1.2793	4.0727	2.9192	2.3042	2.6995	2.4726	4.8139
v_3	1.9046	18.29	4.0186	5.0356	−1.1495	3.1227	0.044872	4.375	7.9624	5.1901	12.007	5.655
v_4	0.0073357	0.0083713	0.0029981	0.010467	0.0026995	0.0077153	−0.038783	0.012318	0.0041625	0.012926	0.0083713	0.013
v_5	134.87	19265	19.538	176320	780860	−17.82	−0.778	−27.966	48.864	−0.099594	7.3974	−11.341
v_6	6.7567	0.37058	0.095807	15.86	5.8442	24.838	4.064	35.285	−0.14434	1	2.0697	1.92
min t/°C	15	25	10	−10	0	15	25	18	20	25	25	20
max t/°C	60	25	70	100	100	55	25	75	50	75	25	50
max w_i	0.46	0.04	0.33	0.51	0.51	0.36	0.1592	0.3574	0.3445	0.3305	0.0015	0.2535
average viscosity residual δ_η/%	0.00	0.04	0.01	−0.07	0.25	0.13	0.24	−0.56	0.00	0.52	0.02	−0.08
SD of viscosity residual $s_{\delta_\eta/\%}$	1.1	0.22	0.92	1.2	4.7	0.73	0.57	4.3	0.55	3.7	0.03	1.42
no. of points in correlation	134	5	172	72	532	84	16	59	61	29	16	62
no. of inconsistent points	1	0	5	2	17	7	0	1	0	1	0	1
references	12, 43, 49, 92, 124, 209	64, 175	6, 43, 125, 133, 170, 207, 209, 226, 275, 281	3, 13, 78, 237, 241, 286	6, 25, 85, 86, 124, 125, 155, 192, 207, 209, 238, 246, 261, 263, 288, 293, 298	63, 77§, 125, 260, 261	29, 64, 112, 230, 232	16, 19, 217, 252	6, 75, 112, 205, 210§, 214, 220§, 261	19, 252	16	6, 209

name	$Cu(NO_3)_2$	$CuCl_2$	$CuSO_4$	$Fe_2(SO_4)_3$	$FeCl_2$	$FeSO_4$	H_2O_2	H_2SO_4	H_3PO_4	HCH_3CO_2	$HCHO_2$	HCl
v_1	45.382	6.9303	−130.52	42.849	−0.29592	27.074	227.72	7.777	5044.2	−0.5661	10.991	7.124
v_2	1.9172	1.8668	4.0392	2.5502	18.533	0.88474	0.0009402	1.2344	−0.017169	−0.21327	0.039543	1.1919
v_3	3.897	7.4786	10.017	7.1619	8.8165	0.86143	−226.89	1.9164	−5029.7	3.4874	−6.9252	1.6648
v_4	0.0083713	0.0060045	0.060995	0.011598	0.0021966	0.0051165	0.04474	0.0056363	0.0083713	0.074792	0.0064144	0.00096271
v_5	9.4796	65.035	−2324.8	1.239	385.52	7322.9	0.26782	46.007	1497.1	1.4946	19.113	22.185
v_6	2.0697	0.15662	9.2724	−0.57468	0.23964	3.9248	3.3224	2.8307	−74.319	27.123	0.37167	1.479
min t/°C	25	20	15	20	18	25	0	−10	20	15	15	10
max t/°C	25	50	60	50	40	75	20	75	25	55	55	42.5
max w_i	0.3331	0.3751	0.4129	0.3009	0.0366	0.2109	1	0.782	0.8188	1	1	0.36
average viscosity residual δ_η/%	−0.04	−0.02	−0.42	0.01	0.02	0.10	−0.09	−0.53	−1.0	−0.48	0.04	0.09
SD of viscosity residual $s_{\delta_\eta/\%}$	0.50	0.91	4.9	0.29	0.04	1.2	0.62	4.3	5.4	4.2	1.4	1.0
no. of points in correlation	10	73	180	35	43	30	46	141	33	228	153	162
no. of inconsistent points	0	0	5	0	2	0	0	17	4	10	0	1
references	63§, 74, 106	6, 64, 72, 74, 115, 172, 182, 209, 219	10, 16, 43, 61, 74, 202, 209, 225, 266, 295	46, 187	141, 220	19, 60, 209	69, 88, 119, 166, 209, 215	35, 89§, 108, 186, 209, 224, 233, 251, 287	52, 66, 70, 71, 209, 258	34, 57, 66, 98, 99, 124, 161, 162	99, 124	9, 38, 83, 92, 111, 114, 120, 189, 209, 222, 236, 279

name	HCN	HNO_3	K_2CO_3	$K_2Cr_2O_7$	K_2HPO_4	K_2SO_4	K_3PO_4	KBr	KCH_3CO_2	$KCHO_2$	KCl	KH_2PO_4
v_1	1.1509	0.000082264	15.804	3.2419	5873.1	−983.76	2.3507	187.46	6.4061	4.8875	6.4883	1358.1
v_2	3.8268	−1.3972	1.8414	1.1035	5.555	0.00019331	0.46935	−0.00068486	3.0168	4.7802	1.3175	3.8539
v_3	−0.38751	0.11962	2.4469	14.602	4.3106	984.52	2.8206	−188.58	3.6581	1.2964	−0.77785	1.6617
v_4	0.0083713	−0.01992	0.0060894	0.00024669	0.096537	0.0038473	0.0097273	−0.0027457	0.013116	0.044035	0.092722	0.012129
v_5	7.385	−1.4448	36.771	5404950	−0.81881	−9.5001	−6.3284	−1.0079	0.36839	−1.0602	−1.3	−1.0516
v_6	2.0697	0.90694	3.1555	0.26452	0.051171	2.1916	1.9539	0.47054	−0.45637	1.202	2.0811	4.1301
min t/°C	0	4	19	0.2	20.95	0	19.95	0	15	15	5	19.95
max t/°C	0	25	89	89.3	49.95	89.5	49.95	95	55	55	150	44.95
max w_i	1	0.309	0.36	0.4	0.181	0.155	0.209	0.452	0.625	0.813	0.306	0.198
average viscosity residual δ_η/%	2.2	0.00	0.11	−0.20	0.01	0.15	−0.02	0.03	0.08	−0.06	−0.01	0.59
SD of viscosity residual $s_{\delta_\eta/\%}$	8.0	0.24	0.78	3.8	0.70	1.1	0.36	1.2	0.79	6.6	0.38	11

Table 2. (Continued)

name	HCN	HNO_3	K_2CO_3	$K_2Cr_2O_7$	K_2HPO_4	K_2SO_4	K_3PO_4	KBr	KCH_3CO_2	$KCHO_2$	KCl	KH_2PO_4
no. of points in correlation	12	16	45	43	42	200	42	347	126	135	647	60
no. of inconsistent points	0	0	0	0	0	0	9	6	0	0	7	0
references	151, 209	27, 107, 199, 209, 278	53, 90, 116, 179, 209	43, 109, 144, 148, 149, 228	$47^{\S}$	43, 54, 59, 79, 123, 132, 140, 177, 193, 206, 209, 247, 265, 274	$47^{\S}$, $66^{\S}$	20, 43, 83, 94, 95, 118, 125, 135, 137, 164, 179, 183, 196, 209, 228, 250, 253, 263, 264, 273	66, 124, 162	124, 234	6, 21, 38, 43, 59, 66, 83, 85, 97, 101, 104, 126, 128, 140, 150, 158, 164, 167, 179, 183, 188, 194−196, 198, 209, 238, 247, 263, 268, 273, 299	$47^{\S}$, 50, 176, $184^{\S}$

name	KI	KNO_3	KOH	Li_2SO_4	LiCl	$LiNO_3$	LiOH	$Mg(NO_3)_2$	$MgCl_2$	$MgSO_4$	$MnCl_2$	$MnSO_4$
v_1	4.9108	3.9621	57.433	42.491	19.265	16.905	4017.7	31.394	24.032	23.286	28.172	24.038
v_2	1.1146	0.39973	3.8049	2.2133	1.9701	1.5469	3.1299	2.4805	2.2694	0.99267	2.4694	2.1341
v_3	−1.2651	4.259	2.6226	6.064	1.6661	0.63451	14.397	3.7822	3.7108	5.4094	3.8408	6.4724
v_4	0.060535	0.00076988	0.010312	0.0081429	0.01149	0.0083965	0.008346	0.0072105	0.021853	0.0063869	0.0083713	0.0074808
v_5	0.52054	925.92	5219	24.167	−0.79691	156.27	669.91	1195.6	−1.1236	63.574	7.385	0.67399
v_6	0.09659	0.33733	10.946	2.96	−0.017456	4.3685	2.1575	31.967	0.14474	1.6689	2.0697	−0.53884
min t/°C	5	15	−14.1	5	0	0	20	0	15	15	25	20
max t/°C	95	60	40	128.58	95	110	40	50	70	150	25	80
max w_i	0.627	0.495	0.519	0.26	0.454	0.671	0.113	0.37	0.386	0.303	0.42	0.364
average viscosity residual δ_η/%	−0.02	0.01	0.71	0.02	0.23	0.39	−0.02	0.12	0.18	0.17	−0.04	0.05
SD of viscosity residual $s_{\delta_\eta/\%}$	1.4	0.91	2.7	0.67	2.1	3.2	5.0	1.8	3.5	1.1	0.25	2.2
no. of points in correlation	241	146	50	179	438	104	29	212	318	180	14	93
no. of inconsistent points	4	3	0	0	105	25	0	23	32	13	0	15
references	43, 62, 94, 142, 164, 179, 183, 196, 223, 228, 250, 253, 254	22, 43, 63, 73, 125, 136, $148^{\S}$, 179, 194, 199, 204, 209, 241, 242, 263, 270	7, 110, 116, 145, 173, 198, 209, 239, 243, 259, 277	1, 39−42, 141, 201, 294	25, 62, 85, 100, 125, 131, 164, 179, 195, 196, 250, 263, 264, 282, 291, 293	14, 30, 31, 241, 293, 300	110, $117^{\S}$, 239, 259, 272	63, 115, 138, 241, 256, 263, 290	6, 23, 29, 44, 79, 85, 125, 140, 177, 207, 209, 213, 238, 246, 264, 274, 281	16, 17, 43, 44, 67, 79, 80, 115, 125, 134, 140, 150, 177, 179, 182, 206, 209, 228, 248, 263	112, 115, 211, 220, 230, 279	18, 19, 58, 115, 187, 224, 252

name	Na_2CO_3	Na_2HPO_4	$Na_2S_2O_3$	Na_2SO_3	Na_2SO_4	Na_3PO_4	NaBr	$NaCH_3CO_2$	NaCl	$NaClO_3$	NaF	NaH_2PO_4
v_1	16.179	99.53	19.509	0.000044078	26.519	24.444	13.029	13.239	16.222	13.697	158.35	3.9294
v_2	0.48606	2.7075	1.5231	−2.2823	1.5746	0.63525	1.7478	1.6335	1.3229	0.37882	1.6418	0.36692
v_3	1.6033	0.23625	2.7793	5.5871	3.4966	12.95	0.60413	5.6914	1.4849	−1.1782	5.1181	3.4152
v_4	0.01761	−0.017106	0.015159	0.01463	0.010388	0.0025154	0.010804	0.020441	0.0074691	0.0015768	0.0014974	0.0097606
v_5	−6.9093	−0.99535	2.01403E+12	−0.2519	106.23	1639040	17.681	23994	30.78	4065.9	0.42269	3.2217
v_6	2.2356	0.0060289	49.214	4.7169	2.9738	1.5312	2.3831	14.214	2.0583	2.1278	6.1895	0.53556
min t/°C	20	20	20	25	15	19.95	5	25	5	25	5	20
max t/°C	90	50	50	40	150	49.95	60	55	154	55	55	50
max w_i	0.308	0.099	0.575	0.06	0.331	0.076	0.54	0.456	0.264	0.583	0.032	0.3
average viscosity residual δ_η/%	−0.79	0.01	0.16	0.00	0.09	0.00	−0.15	0.07	0.06	0.41	0.03	0.18
SD of viscosity residual $s_{\delta_\eta/\%}$	5.0	1.7	4.2	0.32	0.57	0.26	1.2	1.6	0.48	6.0	0.35	0.69
no. of points in correlation	51	41	26	20	222	35	217	20	552	34	58	53
no. of inconsistent points	0	0	0	0	3	0	0	0	17	0	3	3

Table 2. (Continued)

name	Na_2CO_3	Na_2HPO_4	$Na_2S_2O_3$	Na_2SO_3	Na_2SO_4	Na_3PO_4	NaBr	$NaCH_3CO_2$	NaCl	$NaClO_3$	NaF	NaH_2PO_4
references	53, 113, 116, 168, 179, 206	47, 96, 260, 297	139, 193, 209, 260	209, 260, 284	44, 54, 59, 79, 80, 82, 91, 125§, 142, 150, 168, 177, 206, 209, 247, 248, 274, 294	47, 260	20, 65, 85, 94, 95, 104, 124, 125, 179, 209, 250, 253, 254, 263, 275, 282	43, 66, 228, 235	38, 44, 59, 79, 80, 85, 97, 104, 126, 130, 141, 146, 147, 150, 158, 159, 164, 170, 178−180, 182, 183, 194, 196, 208, 209, 238, 250, 271, 275, 283, 299	37, 43, 209, 240	62, 94, 154, 177, 179, 203, 247	28, 47, 96, 174, 260, 297

name	NaI	$NaNO_3$	NaOH	NH_3	NH_4Cl	NH_4NO_3	$NiCl_2$	$NiSO_4$	$Pb(NO_3)_2$	$Sr(NO_3)_2$	$SrCl_2$	sucrose
υ_1	12.353	5.5367	440.2	2.2287	12.396	3.5287	272.76	26.187	3939.2	18.338	6.3341	59.669
υ_2	1.7906	1.3221	0.0089764	1.17	1.5039	2.5639	5.5487	1.887	0.00036493	1.3569	1.6291	1.7701
υ_3	−0.22864	1.4038	−423.67	3.4326	−1.7756	0.78627	7.4913	5.3108	−3932.2	2.0865	1.9197	10.201
υ_4	0.0084003	0.014242	0.015949	0.0085807	0.23471	0.0095033	0.0062831	0.010252	0.0042192	0.0083713	0.008781	0.010025
υ_5	43.458	0.43102	107.6	54.787	−2.7591	0.65	5.4278	−0.31459	437.27	7.385	−4.265	2716.5
υ_6	3.1429	−0.12645	4.6489	0.84697	2.8408	−0.48778	−0.70825	0.41465	2.1463	2.0697	2.5302	4.8553
min t/°C	5	10	12.5	19.85	10	15	20	15	25	25	10	15
max t/°C	97.83	60	70	39.85	73.5	60	50	60	50	25	72.9	55
max w_i	0.629	0.552	0.56	0.62	0.324	0.785	0.379	0.353	0.375	0.25	0.454	0.25
average viscosity residual δ_η/%	0.04	0.01	0.38	−0.21	0.07	0.14	0.19	−0.04	0.03	0.01	−0.13	0.02
SD of viscosity residual $s_{\delta_{\eta/\%}}$	2.1	2.2	2.7	2.1	0.30	1.2	1.5	1.1	3.9	0.07	3.3	0.23
no. of points in correlation	166	255	174	25	265	277	75	99	14	7	170	81
no. of inconsistent points	0	1	9	0	9	24	1	0	0	0	2	0
references	4, 62, 68, 94, 164, 167, 179, 183, 242, 244, 250§, 253, 254, 282	2, 22, 43, 63, 125, 130, 143, 179, 199, 204, 209, 227, 241, 242, 269, 275	8, 110, 114, 116, 120, 152, 153, 168, 169, 179, 198, 206, 209, 239, 243, 259, 285§	24, 84, 163, 209, 231§, 243§	12, 43, 56, 87, 94, 115, 124, 125, 136, 142, 179, 180, 182, 197, 202, 209, 231, 242	5, 31−34, 43, 87, 209, 180, 240, 241, 255, 289	6§, 64, 76, 112, 200, 209, 212, 219, 229, 252, 261, 262	126, 209, 216, 252	43, 63, 115, 228	63, 172, 263	6, 115, 125, 179, 197, 207, 218, 246, 275	124, 183, 249

name	$ZnCl_2$	$ZnSO_4$
υ_1	12.697	13.593
υ_2	2.8245	1.354
υ_3	2.8195	10.556
υ_4	0.0083713	0.0036034
υ_5	7.385	613.22
υ_6	2.0697	0.19583
min t/°C	25	15
max t/°C	25	55
max w_i	0.52	0.318
avg viscosity residual/mPa•s	0.26	0.00
avg viscosity resitual δ_h/%	0.60	1.8
SD of viscosity residual $s_{\delta_{\eta/\%}}$	13	46
no. of inconsistent points	0	138
references	112, 209, 221, 230, 292	11, 19§, 115, 126, 209, 225, 267§

[a] For references marked with the § symbol, see the Supporting Information for additional notes or qualifiers.

The experimental solution viscosities in Figure 1 fall almost on a line. The calculated viscosities of $CaCl_2$ in Figure 2 show much more spread. This spread is attributed to a combination of experimental uncertainties within each data set and between the various data sets. For example, at 50 °C and close to a mass fraction of 0.3, data points from four references are available.[6,25,288,292] The reported solution viscosities are respectively (1.851, 1.72, 1.992, and 1.92) mPa•s, giving $CaCl_2$ viscosities of (31, 25, 40, and 37) mPa•s.

The following model was found to be capable to adequately represent the viscosities of solutes:

$$(\eta_i/\mathrm{mPa{\cdot}s}) = \exp\left(\frac{\upsilon_1(1 - w_\mathrm{w})^{\upsilon_2} + \upsilon_3}{(\upsilon_4(t/°\mathrm{C}) + 1)(\upsilon_5(1 - w_\mathrm{w})^{\upsilon_6} + 1)}\right) \quad (12)$$

or, in logarithmic form

$$\ln(\eta_i/\mathrm{mPa{\cdot}s}) = \frac{\upsilon_1(1 - w_\mathrm{w})^{\upsilon_2} + \upsilon_3}{(\upsilon_4 t/°\mathrm{C} + 1)(\upsilon_5(1 - w_\mathrm{w})^{\upsilon_6} + 1)} \quad (13)$$

where υ_1 to υ_6 are dimensionless empirical constants.

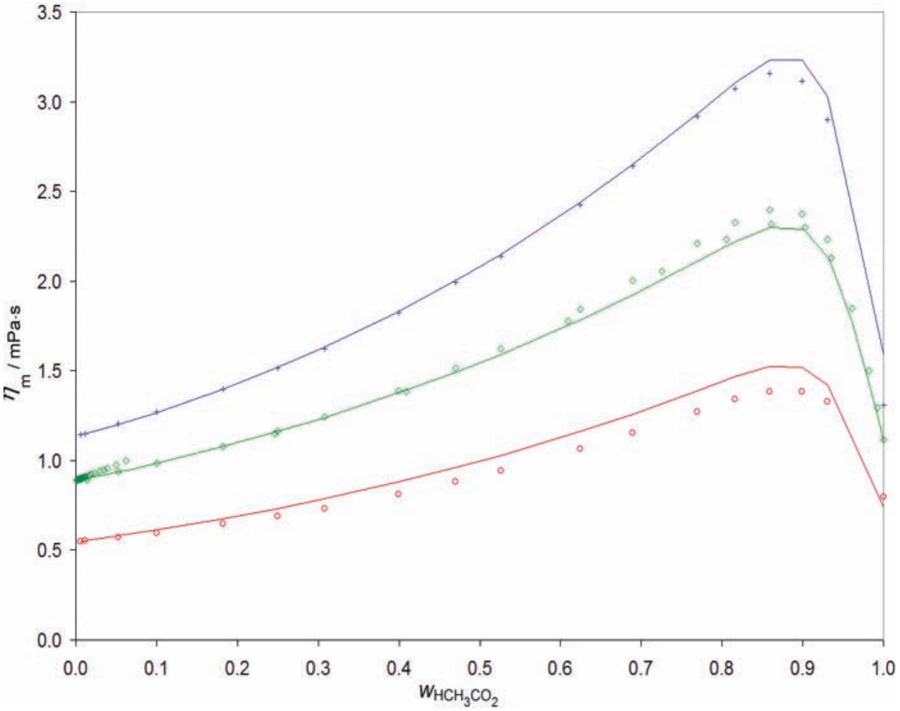

Figure 3. Viscosity of acetic acid aqueous solutions at various temperatures: blue +, experimental at 15 °C; blue line, calculated at 15 °C; green ◇, experimental at 25 °C; green line, calculated at 25 °C; red ○, experimental at 50 °C; red line, calculated at 50 °C.

Table 3. Difference δ_η between Experimental and Calculated Viscosities of Solution of $CaCl_2$ in Water When Fitting over the Entire Concentration Range, When Interpolating over Concentration, and Where Extrapolating over Concentration

	all data: constants v_1 to v_6 fitted over entire data		interpolation: constants v_1 to v_6 fitted only where $w_i < 0.16$ or $w_i \geq 0.40$		extrapolation: constants v_1 to v_6 fitted only where $w_i < 0.32$	
range	residual δ_η/%	SD of residual s_{δ_η}/%	residual δ_η/%	SD of residual s_{δ_η}/%	residual δ_η/%	SD of residual s_{δ_η}/%
$w_{CaCl_2} < 0.08$	3.24	1.83	1.55	0.85	0.47	0.84
$0.08 \leq w_{CaCl_2} < 0.16$	−1.03	1.87	1.69	1.84	0.15	1.74
$0.16 \leq w_{CaCl_2} < 0.24$	1.15	2.10	2.80	1.93	0.10	1.80
$0.24 \leq w_{CaCl_2} < 0.32$	0.54	4.08	0.93	4.17	−0.04	3.37
$0.32 \leq w_{CaCl_2} < 0.40$	0.75	4.21	0.73	4.36	8.46	9.13
$0.40 \leq w_{CaCl_2} < 0.48$	−0.96	4.44	−0.98	4.47	13.20	17.70
$0.48 \leq w_{CaCl_2} < 0.53$	0.31	2.52	0.18	2.44	24.60	24.14

Table 4. Difference δ_η between Experimental and Calculated Viscosities of Solution of $CaCl_2$ in Water When Fitting over the Entire Temperature Range, When Interpolating over Temperature, and Where Extrapolating over Temperature

	all data: constants v_1 to v_6 fitted over entire data/mPa·s		interpolation: constants v_1 to v_6 fitted only where $t <$ (30 or 70) °C $\leq t$/mPa·s		extrapolation: constants v_1 to v_6 fitted only where $t <$ 50 °C/mPa·s	
range	residual δ_η/%	SD of residual s_{δ_η}/%	residual δ_η/%	SD of residual s_{δ_η}/%	residual δ_η/%	SD of residual s_{δ_η}/%
0 °C ≤ t < 10 °C	−1.00	2.53	−0.47	2.31	−0.69	2.36
10 °C ≤ t < 20 °C	−0.10	4.92	0.17	4.95	0.05	4.92
20 °C ≤ t < 30 °C	0.46	4.13	0.41	4.03	0.39	4.10
30 °C ≤ t < 40 °C	0.67	5.24	0.52	5.57	0.39	5.30
40 °C ≤ t < 50 °C	1.38	5.90	1.28	6.32	0.84	6.01
50 °C ≤ t < 60 °C	1.00	6.63	0.67	7.69	0.11	7.31
60 °C ≤ t < 70 °C	−1.33	5.27	−2.16	5.80	−2.86	5.77
70 °C ≤ t < 80 °C	−0.14	5.70	−0.74	5.89	−2.08	5.95
80 °C ≤ t < 90 °C	−0.35	8.26	−0.64	7.96	−2.72	7.96
90 °C ≤ t < 100 °C	−2.21	10.99	−1.76	10.47	−5.06	10.32

The lines in Figures 1 and 2 show the fitted solution and $CaCl_2$ viscosities, respectively. The fit for the solution viscosity is excellent, but the $CaCl_2$ viscosity is being underestimated at low concentration. However, the solute viscosity impact on the solution viscosity decreases as the solute concentration tends toward 0; therefore, this underestimation does not affect the quality of the fit for the solution viscosity.

Results

A nonlinear least-squares method was used to evaluate the terms v_1 to v_6 in eq 12 for all 73 solutes studied herein for

Table 5. Results for Viscosities of Solutions of More than One Solute in Water[a]

	$(NH_4)_2SO_4$ (1) KBr (2) NaCl (3)	$(NH_4)_2SO_4$ (1) KCl (2)	$(NH_4)_2SO_4$ (1) Na_2SO_4 (2)	$(NH_4)_2SO_4$ (1) $NiSO_4$ (2)	$CuCl_2$ (1) HCl (2)	$Fe_2(SO_4)_3$ (1) $FeSO_4$ (2) H_2SO_4 (3)	$Fe_2(SO_4)_3$ (1) K_2SO_4 (2)	$Fe_2(SO_4)_3$ (1) KBr (2)
avg viscosity residual δ_η/%	−0.24	−0.61	−2.6	−0.60	−18	−18	−4.8	−2.7
SD of viscosity residual s_{δ_η}/%	1.1	0.37	1.7	0.62	11	34	3.4	3.0
no. of points used in correlation	12	6	6	33	24	112	84	91
no. of inconsistent points	0	0	0	0	0	0	0	0
min t/°C	25	25	25	10	30	25	20	20
max t/°C	25	25	25	35	90	80	50	50
max w_1	0.15	0.11	0.24	0.04	0.17	0.22	0.17	0.17
max w_2	0.22	0.06	0.10	0.04	0.21	0.15	0.05	0.11
max w_3	0.12					0.07		
references	92	92	92	185	102.§ 279	190§	45	48

	$BaCl_2$ (1) NaCl (2)	$Ca(NO_3)_2$ (1) NaCl (2)	$CaCl_2$ (1) KCl (2)	$CaCl_2$ (1) NaCl (2)	$Fe_2(SO_4)_3$ (1) KCl (2)	$Fe_2(SO_4)_3$ (1) KNO_3 (2)	$Fe_2(SO_4)_3$ (1) Na_2SO_4 (2)	$Fe_2(SO_4)_3$ (1) NaBr (2)
avg viscosity residual δ_η/%	0.17	−43	−0.32	1.3	−3.0	−4.9	−5.3	−3.5
SD of viscosity residual s_{δ_η}/%	0.56	55	2.5	4.1	2.9	5.0	3.2	3.1
no. of points used in correlation	14	18	118	114	91	112	63	112
no. of inconsistent points	0	0	0	0	0	0	0	0
min t/°C	20	25	25	25	20	20	20	20
max t/°C	20	25	25	25	50	50	50	50
max w_1	0.13	0.36	0.27	0.26	0.17	0.17	0.17	0.17
max w_2	0.08	0.14	0.18	0.15	0.07	0.09	0.12	0.09
references	92, 170	191	157, 298	156, 191, 192, 245, 298	45	48	46§	48

	$Fe_2(SO_4)_3$ (1) NaCl (2)	$Fe_2(SO_4)_3$ (1) $NaNO_3$ (2)	HCl (1) KCl (2) NaCl (3)	KCl (1) $MgCl_2$ (2) $MgSO_4$ (3) NaCl (4)
avg viscosity residual δ_η/%	−3.2	−3.8	0.44	0.23
SD of viscosity residual s_{δ_η}/%	4.6	3.5	0.80	0.31
no. of points used in correlation	84	112	29	35
no. of inconsistent points	0	0	0	0
min t/°C	20	20	25	25
max t/°C	50	50	25	150
max w_1	0.17	0.17	0.10	0.002
max w_2	0.10	0.08	0.19	0.009
max w_3			0.15	0.010
max w_4				0.077
references	46	48	92	81

	KCl (1) NaCl (2)	KCl (1) Sucrose (2)	LiCl (1) $LiNO_3$ (2)	$MgSO_4$ (1) NaCl (2)	Na2SO4 (1) NaCl (2)	NaCl (1) NH_4NO_3 (2)	NaCl (1) sucrose (2)
avg viscosity residual δ_η/%	1.8	−2.0	6.5	−0.23	0.20	−5.1	−1.0
SD of viscosity residual s_{δ_η}/%	4.8	1.4	25	1.0	0.24	7.4	1.1
no. of points used in correlation	180	9	40	75	63	17	14
no. of inconsistent points	0	0	0	0	0	0	63
min t/°C	25	25	10	25	25	25	25
max t/°C	150	25	80	150	150	25	25
max w_1	0.21	0.015	0.32	0.15	0.03	0.22	0.011
max w_2	0.18	0.20	0.18	0.15	0.17	0.39	0.20
references	59, 81, 92,§ 158, 191, 299	183,§ 249, 296	127	80, 81, 191	59, 80, 81	191	26,§ 183,§ 249, 296

[a] For references marked with the § symbol, see the Supporting Information for additional notes or qualifiers.

the 9000 data points that were used in these calculations. The methodology is very similar to the one used in our previous paper:

1. Initial values were entered for ν_1 to ν_6.
2. A residual δ_η was calculated by subtracting the experimental solution viscosity from its calculated value ($\delta_\eta = \eta_m - \eta_{calc}$).
3. The sum of the square of the residuals δ_η was calculated, and this value was minimized by varying ν_1 to ν_6.
4. The data were checked for consistency (see below). If an inconsistent datum was found, it was removed, and steps 2 and 3 were repeated. This was repeated until there was no more inconsistent data.

As in our previous work, experimental data points with significant error were removed from the calculation of the constants. Significant error here is defined as a point where the residual is greater than the average residual ± 4 times the standard deviation of the residuals. This rule was not blindly followed. If the residual could be seen as being part of a pattern of residuals going steadily worse, typically as the solute mass fraction was going toward a minimum or a maximum, the point was kept. However, if no pattern was found it was removed. This is usually a sign of a measurement or a transcription error. All inconsistent data points have been kept in the Supporting Information for further study and are identified by an exclamation point "!" to the right of the residuals.

The values of constants υ_1 to υ_6 from eq 12 are given in Table 2. All solutes are presented in alphabetical order and are referred by their chemical formula, except for sucrose. The table also includes for each solute the number of consistent points n, the number of inconsistent points, the average viscosity residual δ_η, and its standard deviation $s_{\delta_\eta} = \sqrt{(\sum \delta_\eta^2)/(n-1)}$. The average residual δ_η for all consistent points $\sum\delta_n/n$ is 0.05 % with a standard deviation s_{δ_η} of 3.7 % of the measured solution viscosity.

The fit is generally excellent. As an example, Figure 3 shows the mixture viscosity for the system water acetic acid, over its entire concentration range and at temperatures of 15 °C, 25 °C, and 50 °C.

Interpolation and Extrapolation of Equation 12. Two essential properties of models, if they are to be useful in real life applications, are their ability to interpolate and extrapolate outside of the available experimental data. Polynomial models are notoriously bad in this respect. Also difficult to use are models where a set of constants is given for each experimental temperature. Equation 12 was tested for its robustness in interpolating and extrapolating on concentration and on temperature. $CaCl_2$ was again chosen as the test case.

As shown in Table 3, there is no statistical difference in the results when interpolating data on concentration. Extrapolation is however different. Extrapolating by 10 % to 20 % over the maximum concentration where data are available is probably acceptable for many applications. Above this, the error increases rapidly and the predicted viscosity is lower than its actual value. [Except for cases where the solute decreases the viscosity of the solution, in which case the predicted solution viscosity will be higher than its actual value.] Note that conversely the fit on the reduced range is better than when fitting over the entire data range.

$CaCl_2$ is a particularly difficult solute to model. Many solutes show a much flatter viscosity−composition curve, and extrapolation over composition would probably be acceptable over a wider range than in the case studied here.

Table 4 gives the results for interpolation and extrapolation on temperature. In this case, there is no statistical difference between the results based on fitting the parameters of eq 12 on the complete data set, on interpolating the data, or on extrapolating the data by 50 °C.

Prediction of the Viscosity of Solutions of More Than One Solute in Water. Equation 12 can be used to predict the viscosity of solutions containing an arbitrary number of solutes. The effect of solutes at low concentration (say $w_i < 0.01$) is minor and often negligible. However, if many solutes are present at moderate concentration, the mixture viscosity is significantly affected, and eq 12 will correctly predict this behavior.

Numerous data sets of solutions of more than one solute in water were found. Table 5 presents some results for 27 systems and 1688 data points. The average viscosity residual δ_η is −2.7 % (standard deviation s_{δ_η} 16.1 %) of the measured solution viscosity. This does not seem very good, but upon close examination the high standard deviation is driven in large part by some outliers at high concentration, notably in the system LiCl and $LiNO_3$ as measured by Iyoki,[127] where the residual viscosities at very high concentration are as large as 5.6 mPa·s (18 % of the measured viscosity), and to a lesser extent in the system $Ca(NO_3)_2$ and NaCl as reported by Nowlan[191] where the maximum residual viscosity at high concentration is −3.5 mPa·s (−100 % of the measured viscosity). The median standard deviation s_{δ_η} for all data sets is 3.5 % of the measured viscosity, which is much better and comparable to the standard deviation of the systems with one solute only.

In general, as shown in the median results, eq 12 will predict the solution viscosity with sufficient accuracy for most calculations. In some systems, such as KCl, $MgCl_2$, $MgSO_4$, and NaCl as reported by Fabuss[79] and HCl, KCl and NaCl as reported by Goldsack,[92] there is no significant difference between the experimental and calculated values (standard deviation of 0.4 % and 0.8 % of the solution average viscosity, respectively).

Conclusion

This review proposes a mixing rule and a viscosity model that are applicable to solutions of an arbitrary number of solutes in water, with no limits on solute concentration or solution temperature. The validity of the mixing rule and viscosity model was demonstrated by predicting the viscosity of solutions of up to four solutes with a median standard deviation of 3.5 % of the viscosity, which is more than adequate for most engineering calculations.

The viscosity model can be used with confidence when interpolating the solute concentration or the solution temperature over the range of the published data. Extrapolating over temperature, even by as much as 50 °C, also gives good results. However, trying to calculate the solution viscosity when the solute concentration is higher than the published data does not give good results, especially when the viscosity increases steeply with the concentration. Note should therefore be taken of the data's maximum concentration tabulated in Table 2, and calculations above this concentration should be interpreted with caution.

Finally, the viscosity model works well at moderate pressure (up to 1 MPa), but is subject to a small but systematic error as the pressure increases, underestimating the solution viscosity. This underestimation increases as the solution pressure increases but probably remains acceptable for engineering calculations with pressures as high as 10 MPa or more.

Supporting Information Available:

Calculation spreadsheets for all the solutions presented in Tables 2 and 5; density data for all solutes reviewed in this publication, using the same methodology as in our previous publication;[160] file _PropertiesAqueousSolution.xls containing a summary of all the viscosity and density data plus a simple Visual Basic program that allow new functions to be used in Excel in order to calculate a solution density or viscosity; file _Read_Me_First.txt for more details; and density data for $NaHCO_3$ updated[113,179,206,231,280] and included for reference even if no viscosity data are included as supplementary information. This material is available free of charge via the Internet at http://pubs.acs.org.

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