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The Viscosity of Aqueous Alkali-Chloride Solutions up to 623 K, 1,000 bar, and High Ionic Strength

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Abstract An accurate viscosity (dynamic viscosity) model is developed for aqueous alkali-chloride solutions of the binary systems, LiCl–H₂O, NaCl–H₂O, and KCl– H2O, from 273 K to 623 K, and from 1 bar to 1,000 bar and up to high ionic strength. The valid ionic strengths for the LiCl–H₂O, NaCl–H₂O, and KCl–H₂O systems are 0 to 16.7 mol · kg⁻¹, 0 to 6 mol · kg⁻¹, and 0 to 4.5 mol · kg⁻¹, respectively. Comparison of the model with about 4,150 experimental data points concludes that the average absolute viscosity deviation from experimental data in the above range is within or about 1 % for the LiCl–H₂O, NaCl–H₂O, and KCl–H₂O mixtures, indicating the model is of experimental accuracy. With a simple mixing rule, this model can be extrapolated to predict the viscosity of ternary aqueous alkali-chloride solutions, making it useful in reservoir fluid flow simulation. A computer code is developed for this model and can be obtained from the author: (maoshide@cugb.edu.cn).

Keywords Aqueous alkali-chloride solutions · KCl · LiCl · NaCl · Viscosity · Water

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List of symbols

1 Introduction

Aqueous electrolyte solutions play an important role in natural and industrial processes, and an understanding of their thermophysical properties is of great interest in chemical engineering and geochemical applications. One of the most important thermophysical properties is the viscosity as a function of temperature, pressure, and salt concentration. The viscosity has been widely used for fluid flow simulations $[1-3]$ $[1-3]$ and engineering designs whenever electrolyte solutions must be stirred, pumped, or moved through pipelines and other equipment. In the study of a sedimentary basin, the viscosity is also important, because variations in viscosity of formation waters have a significant effect on the flow pattern, with consequences for various basin processes.

Therefore, experimentalists have done quite a lot of work in measuring the viscosity of binary aqueous salt solutions since the beginning of the last century. A number of measurements have been reported from numerous laboratories. We have collected the viscosity data of the LiCl–H₂O, NaCl–H₂O, and KCl–H₂O systems as shown in Table [1.](#page-2-0) However, these experimental data are still scattered and cover only a limited temperature–pressure–composition space, and are inconvenient to use. Hence, theorists have devoted extensive efforts to the modeling of the viscosity of these aqueous electrolyte solutions so as to interpolate between the data points or extrapolate beyond the data range $[4-25]$ $[4-25]$.

However, the published models referenced above are found to possess intolerable deficiencies, which led to the motivation of this study. These models either cover a small temperature–pressure range or cannot reproduce existing experimental data. There are many published models of these binary aqueous systems, but here we only discuss a few of the most competitive models. For example, Abdulagatov et al. [\[22\]](#page-12-4) presented a semi-empirical model to calculate the viscosity of the $LiCl-H₂O$ system, covering a temperature–pressure–composition $(T-P-m)$ range of 298 K to 574 K, 1 bar to 400 bar, and 0 to 16.4 mol \cdot kg⁻¹ with a deviation about equal to the experimental uncertainty (1.5 %) from their data. However, this model is inconvenient to use because the parameters of the model are not given as functions of temperature and pressure. For the NaCl–H₂O system, there are two viscosity models covering a large *T* –*P*–*m* region. One model was developed by Spivey et al. [\[18\]](#page-12-5), which is compared with reliable experimental data (273 K to 423 K, 1 bar to 350 bar, and 0 to 6 mol·kg⁻¹)

Reference	Temperature (K)	Pressure (bar)	Molality $(mol \cdot kg^{-1})$	Ν	Quantity measured
LiCl-H ₂ O system					
$[33]$	290.97-298.15	1.01	$0 - 16.68$	31	η_c
$[34]$	298.15	1.01	$0.05 - 1.022$	5	$\eta_{\rm r}$
$[35]$	298.15	1.01	$0.0071 - 0.2201$	18	$\eta_{\rm r}$
$[32]$	298.15	1.01	$0.09 - 4.236$	12	$\eta_{\rm r}$
$[36]$	298.15	1.01	0.726-5.528	10	$\eta_{\rm r}$
$[37]$	298.15	1.01	0.506-4.369	6	$\eta_{\rm r}$
$[38]$	298.15	1.01	0.954-5.618	9	$\eta_{\rm r}$
$[39]$	298.15	1.01	$0.126 - 0.755$	5	$\eta_{\rm r}$
$[40]$	288.15-328.15	1.01	$0.05 - 10$	70	η_{sol}
$[41]$	278.15-368.15	1.01	0.1189-1.2128	60	η_{sol}
$[42]$	298.15-623.15	$20 - 300$	0.238-5.897	140	η_{sol}
$[29]$	298.15-598.15	20-400	$0.48 - 5.90$	215	η_{sol}
$[43]$	298.06-363.40	1.01	10.707-16.734	67	η_{sol}
$[22]$	298.15-574.23	$1 - 400$	0.0236-16.393	391	η_{sol}
$NaCl-H2O$ system					
$[34]$	298.15	1.01	$0.05 - 1.022$	5	$\eta_{\rm r}$
$[21]$	273.15–298.15	1.01	$0.002 - 2.0893$	20	$\eta_{\rm r}$
$[32]$	298.15	1.01	0.093-6.664	14	$\eta_{\rm r}$
$[44]$	298.15	1.01	$0.101 - 5.685$	19	$\eta_{\rm r}$
[45]	303.15-328.15	1.01	$1 - 5$	54	$\eta_{\rm T}$
$[46]$	298.15-423.15	$1.01 - 4.74$	0.0999-3.6024	28	$\eta_{\rm r}$
$[37]$	298.15	1.01	0.506-5.589	8	$\eta_{\rm r}$
$[38]$	298.15	1.01	1.0232-5.604	5	$\eta_{\rm r}$
$[47]$	298.15-323.15	1.01	0.0172-5.4524	44	η_{sol}
$[48]$	291.29–313.88	$1 - 314.7$	$2.6 - 5.614$	249	η_{sol}
$[30]$	374.43-629.49	100-1500	$0.91 - 4.04$	78	η_{sol}
$[49]$	291.15-427.15	$1 - 313.9$	0.4775-5.4054	726	η_{sol}
[50]	293.15-623.15	$20 - 300$	0.173-4.278	186	η_{sol}
$[51]$	298.15-623.15	$20 - 300$	0.173-4.278	140	$\eta_{\rm r}$
$[40]$	288.15-328.15	1.01	$0.05 - 5$	56	η_{sol}
$[41]$	278.15-368.15	1.01	$0.1 - 1.2$	60	η_{sol}
$[52]$	297.15-474.65	$1 - 321$	1.0661-6.038	388	η_{sol}
$[53]$	293.15-323.15	1.01	$0.1 - 4.431$	63	η_{sol}
$[54]$	298.15	1.01	0.0234-6.0216	54	η_{sol}
$[55]$	273.15-278.15	$1 - 300$	$0.324 - 0.856$	41	η_{sol}
$[31]$	303.15 - 333.15	$100 - 200$	1.717	8	$\eta_{\rm sol}$
$[56]$	308.15-323.15	1.01	$0.0625 - 1$	15	η_{sol}

Table 1 Viscosity measurements for aqueous LiCl, NaCl, and KCl solutions

Reference	Temperature (K)	Pressure (bar)	Molality (mol \cdot kg ⁻¹)	\boldsymbol{N}	Quantity measured
$KCl-H2O$ system					
$[34]$	298.15	1.01	$0.05 - 1.033$	5	$\eta_{\rm r}$
$[57]$	298.15	1.01	$0.002 - 0.202$	10	$\eta_{\rm r}$
$[58]$	298.15	1.01	$0.507 - 3.266$	4	$\eta_{\rm r}$
$[32]$	298.15	1.01	$0.101 - 4.95$	13	$\eta_{\rm r}$
[59]	298.15-323.15	1.01	$0.102 - 4.649$	18	$\eta_{\rm r}$
[60]	303.15-328.15	1.01	$1 - 5.90$	54	$\eta_{\rm r}$
$[36]$	298.15	1.01	$0.507 - 4.818$	14	$\eta_{\rm r}$
[46]	298.15-423.15	$1.01 - 4.74$	$0.10 - 3.61$	28	$\eta_{\rm r}$
$[37]$	298.15	1.01	0.509-4.587	6	$\eta_{\rm r}$
$[38]$	298.15	1.01	0.4985-3.9925	8	$\eta_{\rm r}$
$[47]$	293.15-323.15	1.01	0.3297-4.5547	45	η_{sol}
[61]	297.65-423.65	$1 - 312.7$	$0.50 - 4.40$	261	η_{sol}
$[40]$	288.15-328.15	1.01	$0.05 - 4$	49	η_{sol}
$[41]$	278.15-368.15	1.01	$0.04 - 1$	69	η_{sol}
[62]	297.55-477.65	$2 - 327$	0.99005-4.6255	331	η_{sol}
[63]	298.15-623.15	$20 - 300$	$0 - 3.3544$	177	η_{sol}
$[53]$	293.15-323.15	1.01	$0.101 - 3.353$	70	η_{sol}
$[54]$	298.15	1.01	$0.02 - 4.82$	24	η_{sol}

Table 1 continued

N, number of measurements; η_{sol} , viscosity of solution; η_r , relative viscosity (η_{sol}/η_{H_2O}) where η_{H_2O} is the viscosity of pure water; η_c , coefficient of viscosity

listed in Table [1,](#page-2-0) indicating that the average absolute deviation is about 2.72 %, well beyond the experimental uncertainty (within or about 1 %). Another viscosity model for the NaCl–H2O system was developed by Kestin et al. [\[6\]](#page-12-7) covering a valid *T* –*P*–*m* range (293 K to 423 K, 1 bar to 350 bar, and 0 to 6 mol·kg⁻¹). This model can reproduce experimental data with an average absolute deviation of 0.55 % in the stated region. However, this model cannot be applied in the low and high temperature regions, e.g., 273 K to 293 K and 423 K to 623 K, where deviations are well beyond the experimental precisions. For the KCl–H2O system, the best model was proposed by Kestin et al. [\[5](#page-12-8)] using 35 parameters with a wide $T-P-m$ range (298 K to 423 K, 1 bar to 350 bar, and 0 to 4.5 mol · kg⁻¹), and the uncertainty of the model is ± 1 %. However, the model cannot predict the viscosity of KCl–H2O solutions from 273 K to 298 K and from 423 K to 623 K.

In order to overcome the deficiencies of the previous models, here we present a simple model to calculate the viscosity of water and binary LiCl–H₂O, NaCl–H₂O, and KCl–H₂O solutions, within or about equal to experimental uncertainties in a large *T* –*P*–*m* range. The framework of the model is presented in Sect. [2,](#page-4-0) and the experimental data are reviewed in Sect. [3.](#page-5-0) Parameterization and comparison with experimental data are shown in Sect. [4.](#page-7-0) Then, in Sect. [5,](#page-7-1) the binary viscosity model is extrapolated to predict the viscosity of ternary aqueous alkali-chloride solutions.

2 Viscosity Model as a Function of Temperature, Pressure, and Composition

The viscosity of aqueous electrolyte solutions depends strongly on temperature, less on salinity, and is much less dependent on pressure. The viscosity of some electrolyte solutions is anomalous. For instance, for the LiCl–H₂O and NaCl–H₂O solutions, the viscosity increases monotonically with the electrolyte concentration, while for the KCl–H₂O system, the viscosity increases slightly up to a maximum value at very low concentrations and low temperatures, then it decreases to a minimum value, and finally it increases monotonically again at higher concentrations. After an extensive search, we find the following model that can accurately correlate the viscosity properties of binary aqueous alkali-chloride solutions, but is much simpler than previous competitive models:

$$
\ln \eta_{\rm r} = Am + Bm^2 + Cm^3 \tag{1}
$$

$$
\eta_{\rm r} = \frac{\eta_{\rm sol}}{\eta_{\rm H_2O}}\tag{2}
$$

where η_r denotes the relative viscosity; η_{sol} refers to the viscosity (Pa · s) of solutions; $\eta_{\text{H}_2\text{O}}$ is the viscosity of pure water in Pa · s; and *m* is the molality (mol · kg⁻¹) of salts (LiCl, NaCl, or KCl). *A*, *B*, and *C* are polynomial functions of temperature *T* (in K):

$$
A = a_0 + a_1 T + a_2 T^2 \tag{3}
$$

$$
B = b_0 + b_1 T + b_2 T^2 \tag{4}
$$

$$
C = c_0 + c_1 T \tag{5}
$$

The parameters (a_i, b_i, c_i) are evaluated from a large number of experimental data as discussed later.

In order to parameterize the model, an accurate viscosity model for liquid H_2O is required. Up to now, a good viscosity model for H_2O is the formulation of the International Association for the Properties of Water and Steam (IAPWS) [\[26](#page-12-9)] with an uncertainty of about 1 % in viscosity, covering a wide $T-P$ range. However, this model is complicated, so we developed a simpler viscosity model for liquid water, as is represented by the following equation:

$$
\ln \eta_{\text{H}_2\text{O}} = \sum_{i=1}^5 d_i T^{i-3} + \sum_{i=6}^{10} d_i \rho_{\text{H}_2\text{O}} T^{i-8} \tag{6}
$$

where the density of pure water, ρ_{H_2O} , is obtained from IAPWS97 [\[27\]](#page-12-10); and parameters *di* are shown in Table [2.](#page-5-1) The average absolute deviation from IAPWS [\[26](#page-12-9)] is 0.24% from $273K$ to $623K$ and from 1 bar to [1,](#page-6-0)000 bar. As can be seen from Fig. 1, the predicted viscosity of water from Eq. [6](#page-4-1) is in excellent agreement with the values calculated from IAPWS [\[26\]](#page-12-9). The accurate viscosity model of water forms the basis for calculating the viscosity of aqueous alkali-chloride solutions.

3 Review of the Viscosity Data of Aqueous Alkali-Chloride Solutions

There are three basic types of viscometers [\[28](#page-13-35)] to measure the viscosity of fluids: capillary, rotational, and moving-body viscometers. When all the necessary precautions are taken, the absolute viscosity of a fluid can be measured to an uncertainty of better than 0.25 %, but the uncertainty is commonly about 1 %. The viscosity measurements of binary aqueous alkali-chloride solutions (LiCl–H2O, NaCl–H2O, or KCl–H2O) are shown in Table [1.](#page-2-0)

3.1 LiCl– H_2O System

Over 1,000 measurements of the viscosity of aqueous LiCl solutions have been reported, with a wide $T-P-m$ range of $278K$ to $623K$, 1 bar to 400 bar, and 0 to 16.7 mol · kg−1. Abdulagatov et al. [\[22\]](#page-12-4) did good experimental work on the system over a wide $T-P-m$ range. By comparison, we find that the data of Akhundov et al. [\[29](#page-13-11)] are inconsistent with those of others in the same regions. Therefore, all experimental data, excluding those of Akhundov et al. [\[29](#page-13-11)], are used in the parameterization that covers a wide $T-P-m$ range of 278 K to 673 K, 1 bar to 400 bar, and 0 to 16.7 mol·kg⁻¹ for the LiCl–H₂O system.

3.2 NaCl–H2O System

The viscosity measurements of aqueous NaCl solutions are the most extensive of all aqueous electrolyte solutions. More than 2,200 data points have been reported. However, experimental data at high pressures are still scarce. Above 350 bar, only Semenyuk et al. [\[30\]](#page-13-18) measured the viscosity of the NaCl–H₂O system. Their data are used as a test of the extrapolation of the model. Most of the experimental data are consistent with each other except for a small number of data points [\[31,](#page-13-26)[32\]](#page-13-3), which apparently deviate from other data sets in the same $T-P-m$ range. So, all data points

Fig. 1 Percentage deviations of this model from IAPWS2009 viscosity formulation [\[26\]](#page-12-9)

but the data points of [\[30](#page-13-18)] and [\[31,](#page-13-26)[32\]](#page-13-3) are taken for the parameterization, with a large range (273 K to 573 K, 1 bar to 350 bar, and 0 to 6 mol·kg⁻¹) for the NaCl–H₂O system.

3.3 KCl–H2O System

About 1,200 experimental data points have been found for the $KCl-H₂O$ system. However, these data are limited to low pressure, and the data at high pressures are still scarce. By comparison, these data are in agreement with each other; hence, all the data are included in the parameterization with the same weight, covering a large range of 278 K to 623 K, 1 bar to 350 bar, and 0 to 4.5 mol \cdot kg⁻¹ for the KCl–H₂O system.

4 Parameterization and Comparison with Experimental Data

The parameters (a_i, b_i, c_i) of Eqs. [3–5](#page-4-2) are fitted directly by linear least-square regression to the experimental viscosity data discussed above. Table [3](#page-7-2) lists the optimized parameters. Although experimental data at high pressures are scarce for the LiCl–H₂O, NaCl–H₂O, and KCl–H₂O systems, this model can be extrapolated beyond the range of data to predict the viscosity of the three systems up to 1,000 bar, because the viscosity of the alkali-chloride solutions increases only very slightly with pressure (see Fig. [2\)](#page-8-0). We compare the data of Semenyuk et al. [\[30](#page-13-18)], whose data are not used for parameterization, with our model for the NaCl– H_2O system, indicating that the model is in agreement with the data up to 1,000 bar and 573 K (average absolute deviation being 2.08 %). The valid temperature range is from 273 K to 623 K and the valid ionic strengths for the LiCl–H₂O, NaCl–H₂O, and KCl–H₂O systems are 0 to 16.7 mol · kg⁻¹, 0 to 6 mol · kg⁻¹, and 0 to 4.5 mol · kg⁻¹, respectively. Compared with over 4,000 data points, the average absolute deviations of the model from the data for the LiCl–H2O, NaCl–H₂O, and KCl–H₂O systems are 1.10 %, 0.73 %, and 1.14 %, respectively.

The average and maximum absolute deviations calculated from this model are listed in Table [4.](#page-9-0) Figures [3,](#page-10-0) [4,](#page-11-0) and [5](#page-11-1) show the viscosity deviations between the experimental results and model predictions for every binary alkali-chloride system. As can be seen from Table [4](#page-9-0) and Figs. [3,](#page-10-0) [4,](#page-11-0) and [5,](#page-11-1) almost all the experimental data are accurately reproduced by this model within or close to the experimental uncertainty.

5 Predicting the Viscosity of Ternary Aqueous Alkali-Chloride Solutions

Due to the experimental data limitation for ternary aqueous alkali-chloride solutions, it is more difficult to fit directly to experimental data than for binary systems. Here,

Parameters	Systems					
	$LiCl-H2O$	$NaCl-H2O$	$KCl-H2O$			
a ₀	$0.62204136 \times 10^{-2}$	-0.21319213	-0.42122934			
a_1	$0.54436974 \times 10^{-3}$	$0.13651589 \times 10^{-2}$	$0.18286059 \times 10^{-2}$			
a ₂	$-0.40443190 \times 10^{-6}$	$-0.12191756 \times 10^{-5}$	$-0.13603098 \times 10^{-5}$			
b ₀	$0.14987325 \times 10^{-1}$	$0.69161945 \times 10^{-1}$	$0.11380205 \times 10^{-1}$			
b ₁	$-0.66617390 \times 10^{-4}$	$-0.27292263 \times 10^{-3}$	$0.47541391 \times 10^{-5}$			
b ₂	$0.52113332 \times 10^{-7}$	$0.20852448 \times 10^{-6}$	$-0.99280575 \times 10^{-7}$			
c ₀	$0.12101624 \times 10^{-5}$	$-0.25988855 \times 10^{-2}$	Ω			
c ₁	$0.17772678 \times 10^{-6}$	$0.77989227 \times 10^{-5}$	Ω			

Table 3 Parameters of Eqs. [3–5](#page-4-2)

Fig. 2 Viscosity of aqueous NaCl solutions up to 1,000 bar

a simple mixing rule [\[19](#page-12-11)] is used to calculate viscosities of ternary systems without ternary parameters;

$$
\eta_{\text{mix}}(T, P, m_t) = \sum_{i=1}^{2} \frac{m_i}{m_t} \eta_i(T, P, m_t)
$$
(7)

where η_{mix} denotes the viscosity of ternary aqueous alkali-chloride solutions, m_i the molality of the binary system, m_t the total molality, and η_i the viscosity of the binary aqueous alkali-chloride solution. Correia et al. [\[19\]](#page-12-11) measured the viscosity of NaCl– KCl–H2O mixture solutions, so these data are compared with the viscosity calculated from Eq. [7](#page-8-1) as shown by Fig. [6.](#page-12-12) It shows that the predicted viscosities are in good agreement with the experimental data of Correia et al. [\[19](#page-12-11)].

Reference	T(K)	P (bar)	m_{KC1} (mol \cdot kg ⁻¹)	\boldsymbol{N}	AAD(%)	MAD(%)
[59]	298.15-323.15	1.01	$0.102 - 4.649$	18	1.288	2.454
[60]	303.15 - 328.15	1.01	$1 - 5.90$	54	0.776	2.154
[36]	298.15	1.01	$0.507 - 4.818$	14	1.660	2.492
[46]	298.15-423.15	$1.01 - 4.74$	$0.10 - 3.61$	28	0.816	2.097
$\left[37\right]$	298.15	1.01	0.509-4.587	6	1.343	2.70
[38]	298.15	1.01	0.4985-3.9925	8	1.627	2.561
[47]	293.15-323.15	1.01	0.3297-4.5547	45	1.018	3.518
[61]	297.65-423.65	$1 - 312.7$	$0.50 - 4.40$	261	1.074	3.874
[40]	288.15-328.15	1.01	$0.05 - 4$	49	1.090	5.043
[41]	278.15-368.15	1.01	$0.04 - 1$	69	0.803	2.744
[62]	297.55-477.65	$2 - 327$	0.99005-4.6255	331	1.388	4.583
[63]	298.15-623.15	$20 - 300$	$0 - 3.3544$	175	1.008	5.553
$\left[53\right]$	293.15-323.15	1.01	$0.101 - 3.353$	70	0.817	3.657
$\left[54\right]$	298.15	1.01	$0.02 - 4.82$	24	1.230	2.471

Table 4 continued

N, number of measurements; *AAD*, average absolute deviations calculated from this model; *MAD*, maximum absolute deviations calculated from this model

Fig. 3 Deviations of this model from experimental data for aqueous LiCl solutions: experimental data are from Table [4](#page-9-0)

6 Conclusions

An accurate viscosity model over a wide range of temperature, pressure, and concentration is developed for the LiCl–H₂O, NaCl–H₂O, and KCl–H₂O systems within experimental uncertainty. The average absolute deviation from extensive experimental viscosity data for the LiCl–H₂O, NaCl–H₂O, and KCl–H₂O systems is 1.10 %,

Fig. 4 Deviations of this model from experimental data for aqueous NaCl solutions: experimental data are from Table [4](#page-9-0)

Fig. 5 Deviations of this model from experimental data for aqueous KCl solutions: experimental data are from Table [4](#page-9-0)

0.73 %, and 1.14 %, respectively. With a simple mixing rule, this model can be extrapolated to predict the viscosity of ternary aqueous alkali-chloride solutions within or near experimental uncertainty. A computer code is developed for this model and can be obtained from the author: (maoshide@cugb.edu.cn).

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Fig. 6 Deviations of this model from experimental data for NaCl–KCl–H₂O solutions

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