Table III. Values of Constants A_{ijk}^0, A_{ijk}^1, and A_{ijk}^2 and $\sigma(V_m^E)$ at 308.15 K

system	A_{ijk}^0, cm^3 mol^{-1}	A_{ijk}^1, cm^3 mol^{-1}	A_{ijk}^2, cm^3 mol^{-1}	$\sigma(V_m^E)$, cm^3 mol^{-1}
methylene bromide (i) + pyridine (j) + β-picoline (k)	−1.388 (±0.1870)	−7.038 (±0.3639)	−82.541 (±13.3)	0.001
pyridine (i) + β-picoline (j) + cyclohexane (k)	−1.405 (±0.0848)	6.0 (±0.29)	−69.327 (±7.56)	0.001
benzene (i) + toluene (j) + 1,2-dichloroethane (k)	−0.344 (±0.04)	0.562 (±0.04)	−5.991 (±0.5)	0.001
benzene (i) + *o*-xylene (j) + 1,2-dichloroethane (k)	0.008 (±0.0002)	2.83 (±0.1)	18.137 (±6.2)	0.002
benzene (i) + *p*-xylene (j) + 1,2-dichloroethane (k)	0.364 (±0.02)	−1.77 (±0.1)	−40.892 (±5.0)	0.002
methylene bromide (i) + pyridine (j)	1.22 (±0.0023)	0.09 (±0.0088)	0.038 (±0.0125)	0.002
methylene bromide (i) + β-picoline (j)	1.3 (±0.0052)	2.69 (±0.0004)	0.138 (±0.0432)	0.002
pyridine (i) + β-picoline (j)	0.017 (±0.0009)	−0.018 (±0.0042)	−0.076 (±0.0129)	0.001
pyridine (i) + cyclohexane (k)	2.36 (±0.0003)	−0.92 (±0.0004)	0.281 (±0.016)	0.002
β-picoline (j) + cyclohexane (k)	2.44 (±0.002)	−1.518 (±0.0112)	1.342 (±0.0084)	0.003

[a] For (i + j), etc.; mixtures A_{ijk}^0 denotes A_{ij}, $A_{ijk}^1 = A_{ij}^1$ and $A_{ijk}^2 = A_{ij}^2$, etc. [b] A_{ij}^n, etc., constants of the (i + j), etc., binary mixtures along with their standard deviations are recorded.

excess volumes for methylene bromide (i) + pyridine (j) + β-picoline (k) and pyridine (i) + β-picoline (j) + cyclohexane (k) ternary mixtures, it increases the molar excess volumes for benzene (i) + toluene (j) + 1,2-dichloroethane (k), benzene (i) + *o*-xylene (j) + 1,2-dichloroethane (k) and benzene (i) + *p*-xylene (j) + 1,2-dichloroethane (k) mixtures.

Glossary

x_i, etc.	mole fraction of i, etc.
$V_m^E(T, x_i, x_j)$	molar excess volumes for ternary (i + j + k) mixture
A_{ijk}^n (n = 0–2)	constants in eq 1
A_{ij}^n (n = 0–2)	constants in eq 1 for (i + j) etc. mixtures.

Registry No. Pyridine, 110-86-1; β-picoline, 108-99-6; cyclohexane, 110-82-7; benzene, 71-43-2; toluene, 108-88-3; 1,2-dichloromethane, 107-06-2; *o*-xylene, 95-47-6; *p*-xylene, 106-42-3.

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Viscosity Studies of Water–Acetamide–Potassium Nitrate Ternary System at Different Temperatures

Marian Woldan

Research and Development Centre for Standard Reference Materials, WZORMAT—Branch in Lodz, 90-132, Lodz, Poland

Viscosities of KNO_3 solutions in water–acetamide mixed solvents were measured within the temperature range 25–85 °C. The values of relative viscosity and coefficient *B* of the viscosity equation of investigated systems were calculated. The effect of KNO_3 and temperature on the structure of the investigated systems has been discussed.

While ion–solvent interactions have been studied in detail in aqueous solutions, less consideration has been given to such effects in nonaqueous solvents but investigations in mixed solvents are scanty. The variation of viscosity with temperature and solvent composition has been employed to study the ion–solvent interaction by many workers (*1*) both in aqueous and nonaqueous solutions. Although it is difficult to obtain information concerning the structural effects from the absolute magnitude of the viscosity, it is possible to obtain more information from the temperature dependence because the structural effects involving hydrogen bonding are relatively weak.

The relative viscosity η_r of a solution can be expressed by the Jones–Dole (*2*) equation

$$\eta_r = \eta/\eta_0 = 1 + Ac^{1/2} + Bc \qquad (1)$$

where η and η_0 are the viscosity of the solution and solvent, respectively, and *A* and *B* are constants depending on solute, solvent, temperature, and pressure.

The coefficient *A* is determined by ion-atmosphere interaction and ionic mobilities. The second parameter *B* in eq 1 has been related to the solute–solvent interaction and it shows strong temperature dependence in water. In a previous paper

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Table I. Density and Viscosity of Aqueous Acetamide Solutions within the Temperature Range 25–85 °C[a]

wt % $AcNH_2$	25 °C		40 °C		60 °C		75 °C		85 °C	
	d	η	d	η	d	η	d	η	d	η
5	1.000 22	0.978	0.995 05	0.710	0.985 58	0.503	0.976 98	0.404	0.970 55	0.354
15	1.006 95	1.186	1.000 81	0.848	0.990 26	0.589	0.981 02	0.461	0.974 22	0.399
30	1.017 15	1.614	1.009 34	1.122	0.996 97	0.757	0.987 26	0.579	0.979 72	0.500
50	1.030 24	2.606	1.019 95	1.694	1.005 70	1.094	0.994 31	0.831	0.986 16	0.703
70					1.011 63	1.711	0.999 60	1.241	0.990 70	1.008
85									0.992 24	1.384
95									0.991 90	1.745
100									0.990 72	2.041

[a] References 6 and 7.

(*3*) we reported measurements of viscosity of water–acetamide–NaI system within the temperature range 25–85 °C. In the present work an attempt has been made to deal with the ion–solvent interaction of KNO_3 in water–acetamide mixed solvents of varying compositions to see the effect of temperature on the B coefficient of the viscosity.

Experimental Section

Acetamide of analytical grade was recrystallized from hot methanol and dried under vacuum over phosphorus pentoxide. Melting point of acetamide was 80–81 °C (*4*). KNO_3 of analytical grade was recrystallized from twice distilled water and dried under vacuum. Solutions were made up by weight. Viscosity measurements were made using Ubbelohde suspended-level viscometers (Schott-Gen Mainz, Jena Glass GDR). These required 10–15 cm^3 of solution and had efflux times of 300–800 s. The design of these viscometers eliminates pressure corrections and minimizes surface tension effects (*5*). The viscometers were calibrated with water and 30% aqueous sucrose solution over the temperature range 20–85 °C by using an equation of the form

$$\eta = d(at - b/t) \tag{2}$$

where η is the dynamic viscosity, d is the density of the liquid, t is the efflux time, and a and b are constants. Calculations with these values showed that the kinetic energy correction b/t was negligible in the present measurements of ratios of closely similar efflux times in the same or in nearly identical viscometers.

The present work was concerned solely with relative viscosities and it was found sufficiently precise to take

$$\eta/\eta_0 = td/t_0d_0 \tag{3}$$

where η, t, and d refer to the solution and η_0, t_0, and d_0 to the solvent. The viscometers were mounted in a thermostated water bath. The mean temperature fluctuation within a measurement was considered to be ±0.02 K. Efflux times were measured with an accuracy of ±0.01 s by means of an electronic timer. Between three and six measurements of each efflux time were taken, achieving an average reproducibility of ±0.1 s. In this work, before a viscometer was filled with a new solution, it was cleaned by flushing 3 times with twice distilled water, then 2 times with methanol, and it was dried. All of solutions were passed through a No. 2 sintered glass filter to trap solid particles. At intervals of about 3 months, the viscometers were filled with freshly prepared and filtered chromic acid. Evaporation of the solvent can cause errors in the molality. They were determined by observing the level change in the lowest viscometer vessel with a cathetometer. These errors are small and they are likely to be much smaller than the total error in B. The densities of solutions were measured by using the magnetic float densimeter (*6*) with precision 1×10^{-5} $g{\cdot}cm^{-3}$.

Results

The results of measurements are given in Tables I and II. The overall uncertainty in the relative viscosity is believed to be of the order $\pm 3 \times 10^{-4}$ and density $\pm 2 \times 10^{-5}$ $g{\cdot}cm^{-3}$. The values of density obtained in this study for aqueous solutions of KNO_3 agree well with the literature data (*8*).

The values of coefficient B were calculated from the Jones–Dole equation in the form

$$(\eta/\eta_0 - 1)c^{1/2} = A + Bc^{1/2} \tag{4}$$

In this work a study of the effects on the coefficient B of higher order concentration terms was made in the "extended" Jones–Dole equation (*9*). This involved fitting the equation

$$(\eta/\eta_0) - 1 = Ac^{1/2} + Bc + Dc^2 \tag{5}$$

in stages, estimating the coefficients B and D by least squares and then successively reducing the number of data points in the analysis by omitting the point currently at highest concentration, until the value of coefficient D was not significantly different from zero. Equation 1 generally proved satisfactory up to a concentration of 0.15 $mol{\cdot}dm^{-3}$. The values of coefficient A of KNO_3 solutions in water–acetamide mixtures are close to zero (0.003 ± 0.001) and almost do not change with increase in temperature. The values of coefficient B are given in Table III. The mean error was ±0.006 – 0.010 except for acetamide, which have error estimated as ±0.02 $dm^3{\cdot}mol^{-1}$.

Discussion

As it is seen from Table II, at 25 °C the viscosity of KNO_3 solutions in mixed water–acetamide solvents is smaller than the viscosity of solvent for mixed solvents containing less than 50 wt % of acetamide and it decreases with the concentration of KNO_3 in solution. In the case of aqueous acetamide solvents containing 50 wt % $AcNH_2$ at 25 °C and all mixed solvents at 40, 60, 75, and 85 °C the viscosity of KNO_3 solutions is larger than the viscosity of solvent and it increases with the growth of concentration of KNO_3 in solution and contents of acetamide in the mixed solvents. It can be supposed that the increase of viscosity of the discussed systems is caused by the growing volume of solvation shell around ions. It seems probable that in the water–acetamide system the mixed water–acetamide clusters occur like in the formamide–water system (*11*).

At higher temperature the mixed clusters are probably more susceptible to disruption than water clusters because hydrogen bonds among water–acetamide molecules are slightly weaker than H bonds in pure water (*12*, *13*). More free molecules of water and acetamide can take part in ionic solvation of KNO_3.

In the case of water and mixed solvents containing less than 50 wt % $AcNH_2$ at 25 °C the viscosity decreases probably because of the breaking effect of KNO_3 on the structure of the solvent.

Table II. Density and Relative Viscosity of Water-Acetamide-KNO_3 Ternary System within the Temperature Range 25–85 °C

(a) 0 wt % $AcNH_2$/Water

m_{KNO_3}, mol/kg	25 °C		40 °C		60 °C		75 °C		85 °C	
	d	η_r	d	η_r	d	η_r	d	η_r	d	η_r
0.0201	0.998 26	0.9992	0.993 41	1.0004	0.984 38	1.0013	0.975 99	1.0020	0.969 77	1.0023
0.0502	1.000 05	0.9977	0.995 16	1.0005	0.986 08	1.0030	0.977 63	1.0046	0.971 52	1.0055
0.0699	1.001 58	0.9966	0.996 64	1.0005	0.987 56	1.0041	0.979 13	1.0064	0.973 01	1.0076
0.1002	1.003 04	0.9950	0.998 15	1.0006	0.989 01	1.0058	0.980 58	1.0090	0.974 46	1.0108
0.1498	1.006 09	0.9922	1.001 09	1.0006	0.991 90	1.0086	0.983 42	1.0134	0.977 38	1.0161
0.2002	1.009 17	0.9894	1.004 24	1.0006	0.994 82	1.0113	0.986 29	1.0177	0.980 39	1.0213
0.2992	1.015 23	0.9837	1.010 08	1.0005	1.000 65	1.0167	0.991 95	1.0264	0.986 06	1.0317
0.3994	1.021 37	0.9779	1.015 98	1.0006	1.006 44	1.0222	0.997 68	1.0340	0.991 80	1.0422
0.4977	1.027 61	0.9722	1.022 01	1.0006	1.012 26	1.0274	1.003 34	1.0434	0.997 61	1.0525

(b) 5 wt % $AcNH_2$ 95 wt % H_2O

m_{KNO_3}, mol/kg	25 °C		40 °C		60 °C		75 °C		85 °C	
	d	η_r	d	η_r	d	η_r	d	η_r	d	η_r
0.0200	1.001 45	0.9993	0.996 25	1.0004	0.986 75	1.0014	0.978 14	1.0021	0.971 70	1.0024
0.0495	1.003 26	0.9979	0.998 03	1.0006	0.988 46	1.0032	0.979 84	1.0047	0.973 40	1.0056
0.0694	1.004 47	0.9969	0.999 21	1.0007	0.989 61	1.0044	0.980 98	1.0065	0.974 54	1.0078
0.0989	1.006 26	0.9954	1.000 96	1.0009	0.991 32	1.0061	0.982 67	1.0092	0.976 22	1.0110
0.1498	1.009 25	0.9928	1.003 88	1.0010	0.994 16	1.0091	0.985 50	1.0137	0.979 03	1.0164
0.1979	1.012 24	0.9902	1.006 80	1.0012	0.997 00	1.0119	0.988 33	1.0183	0.981 84	1.0217
0.2968	1.018 14	0.9849	1.012 51	1.0014	1.002 53	1.0177	0.993 93	1.0269	0.987 42	1.0323
0.3956	1.023 95	0.9795	1.018 17	1.0015	1.008 02	1.0234	0.999 41	1.0357	0.992 92	1.0430
0.4948	1.029 70	0.9742	1.023 77	1.0017	1.013 52	1.0290	1.004 81	1.0444	0.998 36	1.0535

(c) 15 wt % $AcNH_2$ + 85 wt % H_2O

m_{KNO_3}, mol/kg	25 °C		40 °C		60 °C		75 °C		85 °C	
	d	η_r	d	η_r	d	η_r	d	η_r	d	η_r
0.0201	1.008 17	0.9994	1.001 99	1.0005	0.991 39	1.0015	0.982 14	1.0022	0.975 34	1.0025
0.0497	1.009 96	0.9983	1.003 71	1.0010	0.993 07	1.0035	0.983 79	1.0051	0.977 00	1.0060
0.0699	1.011 15	0.9974	1.004 86	1.0012	0.994 18	1.0048	0.984 88	1.0071	0.978 10	1.0084
0.0999	1.012 96	0.9962	1.006 62	1.0016	0.995 90	1.0068	0.986 58	1.0100	0.979 80	1.0118
0.1497	1.015 87	0.9940	1.009 45	1.0021	0.998 67	1.0101	0.989 31	1.0149	0.982 52	1.0176
0.1979	1.018 78	0.9918	1.012 27	1.0026	1.001 43	1.0133	0.992 04	1.0197	0.985 24	1.0233
0.2968	1.024 49	0.9873	1.017 90	1.0035	1.006 91	1.0197	0.997 45	1.0293	0.990 70	1.0347
0.3956	1.030 36	0.9827	1.023 51	1.0043	1.012 41	1.0262	1.002 91	1.0389	0.996 12	1.0462
0.4948	1.036 02	0.9782	1.029 11	1.0052	1.017 88	1.0324	1.008 38	1.0485	1.001 55	1.0574

(d) 30 wt % $AcNH_2$ + 70 wt % H_2O

m_{KNO_3}, mol/kg	25 °C		40 °C		60 °C		75 °C		85 °C	
	d	η_r	d	η_r	d	η_r	d	η_r	d	η_r
0.0200	1.018 30	0.9998	1.010 46	1.0008	0.998 06	1.0018	0.988 33	1.0024	0.980 79	1.0027
0.0494	1.019 98	0.9992	1.012 10	1.0018	0.999 67	1.0040	0.989 90	1.0056	0.982 39	1.0064
0.0693	1.021 12	0.9987	1.013 22	1.0023	1.000 77	1.0056	0.990 98	1.0078	0.983 47	1.0089
0.0991	1.022 83	0.9980	1.014 90	1.0032	1.002 40	1.0079	0.992 59	1.0110	0.985 09	1.0126
0.1494	1.025 66	0.9967	1.017 68	1.0045	1.005 13	1.0118	0.995 27	1.0164	0.987 78	1.0188
0.1981	1.028 48	0.9954	1.020 46	1.0058	1.007 85	1.0155	0.997 94	1.0217	0.990 47	1.0249
0.2967	1.034 04	0.9927	1.025 98	1.0083	1.013 32	1.0231	1.003 27	1.0323	0.995 85	1.0371
0.3957	1.039 60	0.9899	1.031 46	1.0107	1.018 92	1.0306	1.008 63	1.0430	1.001 25	1.0493
0.4946	1.045 20	0.9872	1.037 03	1.0132	1.024 36	1.0380	1.014 02	1.0535	1.006 72	1.0615

(e) 50 wt % $AcNH_2$ + 50 wt % H_2O

m_{KNO_3}, mol/kg	25 °C		40 °C		60 °C		75 °C		85 °C	
	d	η_r	d	η_r	d	η_r	d	η_r	d	η_r
0.0201	1.031 31	1.0004	1.021 00	1.0013	1.006 74	1.0022	0.995 35	1.0028	0.987 21	1.0031
0.0499	1.032 93	1.0006	1.022 60	1.0029	1.008 31	1.0052	0.996 93	1.0066	0.988 79	1.0074
0.0693	1.033 98	1.0007	1.023 64	1.0039	1.009 34	1.0072	0.997 96	1.0091	0.989 83	1.0103
0.0994	1.035 61	1.0008	1.025 26	1.0054	1.010 94	1.0101	0.999 56	1.0129	0.991 43	1.0146
0.1496	1.038 32	1.0009	1.027 94	1.0078	1.013 57	1.0151	1.002 19	1.0193	0.994 12	1.0218
0.1979	1.040 99	1.0010	1.030 62	1.0102	1.016 19	1.0200	1.004 82	1.0255	0.996 77	1.0289
0.2975	1.046 40	1.0011	1.036 10	1.0149	1.021 62	1.0297	1.01018	1.0380	1.002 19	1.0431
0.3962	1.051 87	1.0012	1.041 54	1.0195	1.026 99	1.0394	1.015 58	1.0506	1.007 64	1.0574
0.4952	1.057 33	1.0013	1.046 98	1.0242	1.032 36	1.0490	1.020 96	1.0630	1.013 06	1.0715

(f) 70 wt % $AcNH_2$ + 30 wt % H_2O

m_{KNO_3}, mol/kg	60 °C		75 °C		85 °C	
	d	η_r	d	η_r	d	η_r
0.0205	1.012 67	1.0029	1.000 65	1.0034	0.991 76	1.0037
0.0499	1.014 18	1.0069	1.001 10	1.0081	0.993 29	1.0089
0.0786	1.015 67	1.0095	1.003 68	1.0113	0.994 84	1.0124
0.0999	1.016 82	1.0135	1.004 83	1.0160	0.995 99	1.0176

Table II (Continued)

(f) 70 wt % $AcNH_2$ + 30 wt % H_2O

m_{KNO_3}, mol/kg	60 °C d	60 °C η_r	75 °C d	75 °C η_r	85 °C d	85 °C η_r
0.1499	1.019 43	1.0202	1.007 46	1.0239	0.998 68	1.0263
0.2000	1.022 04	1.0267	1.010 08	1.0318	1.001 37	1.0349
0.3000	1.027 33	1.0399	1.015 42	1.0473	1.006 84	1.0521
0.3977	1.032 62	1.0530	1.020 68	1.0629	1.012 24	1.0693
0.4963	1.037 78	1.0660	1.026 08	1.0784	1.017 58	1.0865

(g) 85 °C

m_{KNO_3}, mol/kg	85 wt % $AcNH_2$ d	85 wt % $AcNH_2$ η_r	m_{KNO_3}, mol/kg	95 wt % $AcNH_2$ d	95 wt % $AcNH_2$ η_r
0.0202	0.994 33	1.0044	0.0201	0.995 50	1.0052
0.0499	0.995 92	1.0107	0.0498	0.997 12	1.0125
0.0711	0.997 01	1.0149	0.0698	0.998 21	1.0175
0.1002	0.998 58	1.0212	0.0995	0.999 92	1.0248
0.1498	1.001 50	1.0317	0.1497	1.002 78	1.0371
0.2002	1.004 49	1.0421	0.2002	1.005 66	1.0494
0.3001	1.010 32	1.0630	0.2998	1.011 84	1.0737
0.3991	1.015 08	1.0838	0.3997	1.016 53	1.0982
0.4984	1.020 40	1.1045	0.4992	1.021 67	1.1225

Table III. Values of Coefficient B for KNO_3 in Water-Acetamide Mixed Solvents ($dm^3 \bullet mol^{-1}$)

		$AcNH_2$							
T	H_2O	5 wt %, 1.6 mol %	15 wt %, 5.1 mol %	30 wt %, 11.5 mol %	50 wt %, 23.4 mol %	70 wt %, 41.6 mol %	85 wt %, 63.3 mol %	95 wt %, 85.3 mol %	$AcNH_2$[a]
25	-0.060	-0.056	-0.048	-0.030	-0.002				
40	-0.004	-0.001	0.006	0.022	0.044				
60	0.052	0.055	0.062	0.073	0.095	0.129			
75	0.084	0.086	0.094	0.104	0.123	0.154			
85	0.102	0.104	0.112	0.120	0.140	0.170	0.206	0.242	0.27

[a] Reference 10.

Similar conclusions about the effect of KNO_3 on the structure of mixed water-acetamide solvents can be drawn from the analysis of the values of coefficient B. As seen from Table III the values of coefficient B for KNO_3 in water-acetamide mixed solvents are negative at 25 and 40 °C for water and 5 wt % $AcNH_2$ solvent. In other solutions they are positive. The values of coefficient B increase with the growing contents of acetamide in the mixed solvent and temperature. From this according to literature (*14*) it follows that KNO_3 breaks the structure of water and the structure of mixed water-acetamide solvents at 25 °C. At 40 °C the structure of water and mixed solvent containing 5 wt % $AcNH_2$ is only disturbed by KNO_3. In other water-acetamide mixed solvents at 40 °C and all mixed solvents at 60, 75, and 85 °C a small ordering effect of KNO_3 can be observed. It can be supposed that in this case the ordering effect connected with ionic solvation of KNO_3 probably is greater than the disordering effect of the structure of the solvent. The influence of the addition of acetamide on coefficient B of KNO_3 is similar to the effect of temperature and it decreases with the increase of temperature. It is known that the increase of temperature causes the disturbance of structure of solvent, so acetamide probaly also causes loosening of water structure. The breaking effect of acetamide decreases with the increase of temperature because the structure of water is more and more disturbed by thermal motions of molecules (dB/dT decreases).

Glossary

η_r	relative viscosity
η	dynamic viscosity of solution, mPa·s
η_0	dynamic viscosity of solvent, mPa·s
A	viscosity coefficient, $(dm^3 \cdot mol^{-1})^{1/2}$
B	viscosity coefficient, $dm^3 \cdot mol^{-1}$
d	density of solution, $g \cdot cm^{-3}$
d_0	density of solvent, $g \cdot cm^{-3}$
t	efflux time of solution, s
t_0	efflux time of solvent, s
c	concentration, $mol \cdot dm^{-1}$
T	temperature, °C
x	mole fraction

Registry No. KNO_3, 7757-79-1; acetamide, 60-35-5.

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