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Marian Woldan*

RELATIVE VISCOSITY AND FREE ENERGY OF ACTIVATION OF VISCOUS FLOW OF KNO₃ SOLUTIONS IN WATER-ACETAMIDE MIXTURES AT DIFFERENT TEMPERATURES

The relative viscosity of $\rm KNO_3$ solutions in water-acetamide mixtures on the concentration, temperature and the composition of the mixture is discussed. The effective flowing volume of $\rm KNO_3$ and the free energy of activation of viscous flow were calculated and the dependence of these magnitudes on concentration, composition of the water-acetamide mixture and temperature is discussed. The conclusion about the disordering effect of $\rm KNO_3$ on the structure of water-acetamide mixtures has been drawn.

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. The effect of electrolyte on the viscosity of a structured solvent is due to three factors:

a) the solvation of the ions accompanied by a viscosity increase associated with the Einstein volume effect ($\eta^{\ E}),$

b) long range ordering by the ionic field which also increases the viscosity of solvent,

c) the so called "breaking" of solvent structure by the solvated ion which decreases viscosity by disrupting H-bonded groupings.

The decrease of viscosity due to disruption of solvent structure is evidently much greater importance for water than for other solvents. In the previous paper [2] the results of measurements of the viscosity of water-acetamide-NaI ternary system and the effect of NaI on the structure of water-acetamide mixtures have been discussed. It is known that KNO3 has a breaking effect on water structure. Thus it seemed interesting what is the influ-

* Research and Development Centre for Standard Reference Materials WZORMAT - Branch in Łódź. [45]

ence of addition of acetamide on this effect. In the present paper an attempt has been made to deal. with the ion-solvent interaction of KNO, is water-acetamide mixtures of varying composition to see the effect of temperature and hydrogen bonding on the temperature coefficient of relative viscosity dn_/dT and effective flowing volume of KNO3. The free energy of activation of the viscous flow of these solutions have also been calculated for interpreting the influence of KNO₃ upon the viscosity of the water-acetamide mixtures as a rate process. 23RUIAR39MET THEMSERTE TA 23RUIATH SOTHATEDWAYENA RE-

Experimental The method of purification of acetamide and KNO3 and the procedure of measurements of viscosity were described earlier [3]. The results of viscosity measurements of KNO3 solutions made at dm^{-3} a series of concentration covering the range 0.02-0.5 mole in the mixture containing 5, 15, 30, 50 and 70 wt% of AcNH, in water at 25, 40, 60, 75 and 85⁰C were presented previously [3].

Results and Discussion

The relative viscosity of KNO, solutions in water-acetamide mixtures is presented in Figures 1-5. As it is seen from Figures 1-5 the relative viscosity of KNO3 solutions in water-acetamide mixtures increases with the rise of concentration of electrolyte except of mixtures containing smaller than 50 wt% of AcNH₂ at 25⁰C. The increase of the contents of acetamide in the mixed solvent causes the rise of relative viscosity similar as the increase of temperature. It can be supposed that the increase of relative viscosity of the discussed systems is caused by growing volume of solvation layers around ions. From papers [4-6] it is follows that the temperature coefficient of relative viscosity d $\eta_{\rm r}/dT$ better illustrates the structural changes taking place in the solution than relative viscosity. In case of the electrolytes ordering solvent structure the d η_r/dT coefficient is negative and it is positive for structure breaking electrolytes. The values of dŋ /dT coefficient calculated analytically for water-acetamide-KNO3 ternary system are given in Table 1.





Fig. 2. Relative viscosity of KNO_3 solutions in water-acetamide mixtures vs concentration of $KNO_3(m)$ and composition of mixture at $40^{\circ}C: 1-H_2O, 2-5 \text{ wt% AcNH}_2, 3-15 \text{ wt% AcNH}_2,$ $4-30 \text{ wt% AcNH}_2, 5-50 \text{ wt% AcNH}_2$



Fig. 1. Relative viscosity of KNO_3 solutions in water-acetamide mixtures vs concentration of $KNO_3(m)$ and composition of mixture at 25°C: 1-H₂O, 2-5 wt% AcNH₂, 3-15 wt% AcNH₂, 4-30 wt% AcNH₂, 5-50 wt% AcNH₂





Fig. 3. Relative viscosity of KNO₃ solutions in water-acetamide mixtures vs concentration of KNO₃(m) and composition of mixture at 60° C: 1-H₂O, 2-5 wt% AcNH₂, 3-15 wt% AcNH₂, 4-30 wt% AcNH₂, 5-50 wt% AcNH₂, 6-70 wt% AcNH₂ Fig. 4. Relative viscosity of KNO_3 solutions in water-acetamide mixtures vs concentration of $KNO_3(m)$ and composition of mixture at 75°C: 1-H₂O, 2-5 wt% AcNH₂, 3-15 wt% AcNH₂, 4-30 wt% AcNH₂, 5-50 wt% AcNH₂, 6-70 wt% AcNH₂



Fig. 5. Relative viscosity of KNO₃ solutions in water-acetamide mixtures vs concentration of KNO₃(m) and composition of mixture at 85°C: 1-H₂0, 2-5 wt% AcNH₂, 3-15 wt% AcNH₂, 4-30 wt% AcNH₂, 5-50 wt% AcNH₂, 6-70 wt% AcNH₂, 7-85 wt% AcNH₂, 8-95 wt% AcNH₂

As it is seen from Table 1 the values of $d\eta_r/dT$ coefficient of investigated system are positive in the whole range of investigated concentrations and compositions of mixed solvent. It is easy to notice that coefficient $d\eta_r/dT$ increases with the rise of concentration of KNO₃ in the solution and it decreases with the increase in temperature and contents of acetamide in the mixture. From this one can conclude that KNO₃ has disordering effect. on the structure of water-acetamide mixtures. This effect increases

Table 1

Water 95 wt% H_0-5 wt% AcNH_ 85 wt% H_0-15 wt% AcNH_ mKN03 75⁰ 25⁰ 85⁰ 250 75⁰ 40⁰ 60⁰ 40⁰ 60⁰ 85⁰ 25⁰ 40⁰ 600 750 850 [mol.kg⁻¹] 1.0 1.0 0.9 0.05 2.2 1.6 1.1 0.9 0.8 2.2 1.6 1.2 0.9 2.2 1.6 1.2 1.8 2.3 1.9 1.7 4.1 3.2 2.3 1.9 1.7 0.10 4.3 3.3 2.4 2.0 4.1 3.2 0.20 8.6 6.7 4.8 4.0 3.4 8.3 6.6 4.8 3.7 3.1 8.0 7.5 4.8 3.8 3.2 7.0 5.4 4.8 12.2 0.30 12.8 10.0 7.2 5.7 5.2 12.2 9.7 9.6 7.0 5.6 4.8 0.40 16.0 13.2 10.2 8.4 7.4 16.3 12.9 9.6 7.5 6.6 16.3 12.9 9.3 7.6 7.0 0.50 20.4 16.4 12.2 9.8 8.6 20.4 15.9 11.5 9.3 8.6 20.4 15.9 11.5 9.4 8.6 70 wt% H_0-30 wt% AcNH_ 50 wt% H_0-50 wt% ACNH_ 70 wt% AcNH_-30 wt% H_0 MKN03 85⁰ 60⁰ 7.5⁰ 85⁰ 60⁰ 25⁰ 40⁰ 60⁰ 750 25⁰ 400 75⁰ 85⁰ [mol.kg⁻¹] 0.05 2.1 1.5 1.0 0.9 0.8 1.8 1.4 1.0 0.8 0.7 0.8 0.8 0.7 0.10 3.8 2.2 1.7 3.6 2.8 2.1 1.8 3.0 1.6 1.6 1.8 1.7 1.6 0.20 7.9 6.1 4.2 3.3 3.0 6.9 5.5 4.1 3.5 3.3 3.6 3.2 2.9 0.30 11.8 9.2 6.4 5.1 4.7 10.4 8.4 6.1 5.2 4.9 5.3 4.8 4.6 0.40 15.8 12.1 8.4 6.7 6.1 13.6 11.0 8.3 7.0 6.6 6.8 6.5 6.2 9.1 0.50 19.6 15.0 10.6 8.6 7.9 17.1 14.0 10.6 8.5 7.6 7.5 6.5

The values of d_h/dI of KNO₃ solution in water-acetamide mixtures $[1.10^{-3} \text{ deg}^{-1}]$

N. M.

Marian Woldan

Relative Viscosity and Free Energy

with the rise of concentration of KNO_3 but it decreases with the rise in temperature and contents of acetamide in the mixture. From this it follows that the influence of addition of acetamide on water - KNO_3 interactions is similar to temperature effect. It is known that the increase of temperature disorders the structure of solution, therefore one can conclude that addition of acetamide to water decreases the disordering effect of KNO_3 on water structure. One can suppose that the molecules of acetamide form with water the mixed associates in which the H-bonds are slightly weaker than hydrogen bonds in pure water [7, 8].

In order to verify the hypothesis that the volume of solvation layers increase with the rise of temperature and contents of acetamide in the mixed solvent the values of effective flowing volume of KNO_3 in water-acetamide mixtures were calculated on the basis of Einstein's equation [9]:

 $\eta_r = 1 + 2.5\phi$

where ϕ is the fraction of volume occupies by one mole of solute particles in solution. Breslau and Miller [10] have assumed that $\Phi = V_{e}$.c where V_{o} is the effective rigid molar volume of flowing and c is molarity. This V, has been strictly defined as the volume which a mole of solute particles occupies when considered. from purely hydrodynamic reasons, as rigid microscopic spheres. In case of electrolytes V, may be regarded as the volume of ions formed from one mole of electrolyte together with their solvation layers [11]. Finally the values of effective molar volume of flowing for KNC, in water-acetamide mixtures were calculated from

 $V_{\rm e} = (\eta_{\rm r} - 1)/2.5 \cdot c$

As it is seen from Figure 6 the effective molar volume of KNO_3 in water - acetamide mixtures decreases a little with the rise of concentration of electrolyte. By extrapolation of V_e to c = 0 were estimated the limiting effective molar flowing volumes, V_e^0 . The values of V_e^0 obtained are collected in Table 2 and presented in Figure 7 and 8 as a function of the composition of mixed solvent and temperature.

The increase of contents of acetamide in mixture causes the gra-

51

(1)

(2)

dual rise of V_e^0 (Figure 7), Similar V_e^0 increases with the rise of temperature (Figure 8). It is proves that the volume of hydrodynamic unit increases with the increase of contents of acetamide and



Fig. 6. The dependence of the effective molar volume of flowing for KNO₃ in water-acetamide mixtures vs concentration of KNO₃ (m) and composition of mixture at different temperatures: 1-H₂O, 2-5 wt% AcNH₂, 3-15 wt% AcNH₂, 4-30 wt% AcNH₂, 5-50 wt% AcNH₂, 6-70 wt% AcNH₂, 7-85 wt% AcNH₂, 8-95 wt% AcNH₂

Table 2

iciny to	Water			5 wt% AcNH ₂			15 wt% AcNH ₂		
[°c]	v2	⊽e ^o	$\overline{v}_{\rm solv}$	v ₂	⊽°e	Vsolv	v ₂	∇ ^o e	V _{solv}
25	38.0	-24.0	-62.0	39.0	-22.4	-61.4	41.0	-19.2	-60.2
40	39.3	-1.6	-40.9	40.4	-0.4	-40.8	42.4	2.4	-40.0
60	40.4	20.8	-19.6	42.0	22.0	-20.0	44.6	24.8	-19.8
75	41.0	33.6	-7.4	42.7	34.4	-8.3	45.6	37.6	-8.0
85	41.1	40.8	-0.3	42.7	41.6	-1.1	45.0	44.8	-0.2
	And and a second second second	30 wt% AcNH ₂		50 wt% AcNH ₂			70 wt% AcNH ₂		
T	30	wt% A	CNH2	50	wt% A	CNH2	70	wt% Ad	ONH ₂
T [°C]	30 V ₂	wt% A	VNH2 Vsolv	50 V2	wt% Ad	VH2	70 V ⁰ 2	wt% Ad \overline{V}_e^0	VR2
T [⁰ C] 25.	30 V ₂ 44.0	wt% A	cNH ₂ V _{solv} -56.0	50 \overline{v}_{2}^{0} 48.0	wt% An Ve -0.8	V _{solv}	70 V ⁰ 2	wt% Ad	vH2 Vsolv
T [⁰ C] 25. 40	30 \overline{V}_2^0 44.0 45.6	wt% A	v _{solv} √-56.0 -36.8	50 \overline{V}_{2}^{0} 48.0 49.0	wt% An Ve -0.8 17.6	-48.8 -31.4	70 V ₂ -	wt% Ad	V _{solv}
T [⁰ C] 25 40 60	30 \overline{V}_2^0 44.0 45.6 47.0	wt% A	√v _{solv} -56.0 -36.8 -17.8	50 \overline{V}_2^0 48.0 49.0 49.9	wt% Ad Ve -0.8 17.6 38.0	-48.8 -31.4 -11.9	70 √2 - 51.2	wt% Ad	VH2 Vsolv - 0.4
T [⁰ C] 25. 40 60 75	30 \overline{V}_2^0 44.0 45.6 47.0 48.0	wt% An	-56.0 -36.8 -17.8 -6.4	50 \overline{V}_2^0 48.0 49.0 49.9 49.8	wt% Ad Ve -0.8 17.6 38.0 49.2	-48.8 -31.4 -0.6	70 √2 - 51.2 50.6	wt% Ac	ŪVH ₂ √V _{solv} 0.4 11.0

The values of \overline{V}_2^0 , \overline{V}_e^0 and \overline{V}_{solv} for KN03 in water-acetamide mixtures [cm³ mol⁻¹]

temperature. The negative values of V_e^0 for KNO_3 at $25^{\circ}C$ one can explain by large breaking effect of KNO_3 on the structure of water-acetamide mixtures. At higher temperature the mixed water-acetamide associates with weaker than in water H-bonds are more susceptible to disruption than water associates. More free molecules which can take part in ionic solvation appear then in the solution. Similar effect on solvation of KNO_3 has acetamide. Therefore one can say that acetamide disorders of water structure.

More informations concerning of interactions ${\rm KNO}_3-$ mixed solvent one can obtain from the dependence of volume of solvation layers around ions on the composition of mixed solvent and temperature. The volume of solvation layers, $\overline{\rm V}_{\rm Solv}$ one can calculate from equation.

 $V_e^0 = \overline{V}_2^0 + \overline{V}_{solv}$

(3)





Fig. 8. The dependence of the limiting effective molar volume of flowing, V₂^o, for KNO₃ in water-acetamide mixtures vs temperature: 1-H₂O, 2-5 wt% AcNH₂, 3-15 wt% AcNH₂, 4-30 wt% AcNH₂, 5-50 wt% AcNH₂, 6-70 wt% AcNH₂ 54

Marian Woldan

where \overline{V}_2^0 is the partial molal volume of electrolyte. The values of \overline{V}_{solv} obtained for KNO₃ in water-acetamide mixtures are given in Table 2.

As it is seen from Table 2 the volume of solvation layers for KNO3 is negative except the mixed solvents containing 70 wt% of AcNH₂ and the mixtures containing 30 and more wt% of AcNH₂ at 85^oC. The values of V_{solv} increase with the rise of contents of AcNH, in mixed solvent and temperature (see Figure 9). Probably it is causes by disordering effect of temperature and addition of acetamide on water structure. From Figure 9 one can estimate the value of temperatures (T_o) for which \overline{V}_{solv} is equal to zero i.e. the KNO₃ is nonsolvated. The temperature T_o depends on composition of water--acetamide mixture and it decreases with the rise of contents of acetamide in the mixture (Figure 10). Therefore one can conclude that in cases when \overline{V}_{solv} is negative the viscosity of mixed solvent around ions $K^+NO_3^-$ summary is smaller than viscosity in bulk of solvent. It proves that in this case KNO3 disorders the structure of water-acetamide mixtures. In case when $\overline{V}_{solv} > 0$, KNO₃ is positive solvated and it causes the increase in viscosity of mixed solvent. The conclusions obtained from the analysis of the function \overline{V}_{solv} = = $f(x_{ACNH_{o}}, T)$ are confirmed by the course of the dependence of volume expansion coefficient & of water-acetamide-KNO, ternary system as a function of the composition of mixture and temperature [12]. According to Eyring's theory concerning laminar flow of liquid [13] the dynamic viscosity of solution can be described by equation

$$\eta = \frac{hN}{V} \exp \frac{\Delta G^*}{RT} = \frac{hN}{V} \exp \frac{-\Delta S^*}{R} \exp \frac{\Delta H^*}{RT}$$
(4)

where h is Planck's constant, N is Avogadro's number, V is molar volume of solvent, R is the gas constant, T is the temperature in Kelvin and ΔG^* , ΔS^* and ΔH^* are free energy, entropy and enthalpy of activation of viscous flow respectively. The values of particular thermodynamic functions of activation of viscous flow were calculated from following formulas:

$$\Delta G^* = RT \ln \left(\frac{\eta V}{hN}\right)$$
(5)
$$\Delta H^* = Rd \ln \eta / d(\frac{1}{T})$$
(6)



The values of free energy of activation of viscous flow for water-acetamide-KNO₃ ternary system [kJ·mol⁻¹] (% - percentage by weight of AcNH₂)

^C KNO ₃ [mol.dm ⁻³]	0%	5%	15%	30%	50%
		a) 25	°c		- 0.2 - 0
0.02	7.00	7.31	7.95	8.99	10.61
0.05	7.00	7.31	7.95	8.98	10.61
0.07	7.00	7.31	7.95	8.98	10.61
0.10	6.99	7.30	7.94	8.98	10.61
0.15	6.98	7.30	7.94	8.98	10.61
0.20	6.98	7.29	7.93	8.97	10.61
0.30	6.96	7.28	7.92	8.97	10.61
0.40	6.95	7.26	7.91	8.96	10.61
0.50	6.93	7.25	7.90	8.95	10.61
		b) 40 ⁰	Pc		91 1 2 - Ta
0.02	6.56	6.86	7.50	8.51	10.05
0.05	н			8.52	
0.07					all and the second s
0.10		6.87		н	10.06
0.15		H (0	н.	н	170.00
0.20					10.07
0.30	e n 1927			8.53	10.08
0.40			7.51	8.54	10.09
0.50	н				10.10

Table 3 (contd)

CKN03	0%	5%	15%	30%	50%	70%
[mol.am]	L.HPDA					-
			c) 60 ⁰ C			
0.02	6.08	6.38	7.00	8.00	9.52	12.24
0.05			u	8.01	9.53	12.25
0.07	6.09	00.1	7.01		н	12.26
0.10		6.39	н	8.02	9.54	12.27
0.15	6.10	6.40	7.02	8.03	9.55	12.29
0.20	6.11		7.03	8.04	9.56	12.31
0.30	6.12	6.42	7.05	8.06	9.60	12.35
0.40	6.14	6.44	7.06	8.08	9.62	12.39
0.50	6.15	6.45	7.08	8.10	9.64	12.42
19.01	45	27.7 d) 75 ⁰ C	00.1		0.07
0.02	5.77	6.06	6.63	7.62	9.18	11.00
0.05	5.78		6.64	7.63	9.19	11.01
0.07	5.79	6.07	6.65		9.20	11.03
0.10	5.80	6.08	6.66	7.64	9.21	11.04
0.15	5.81	6.09	6.67	7.66	9.23	11.06
0.20	5.82	6.10	6.68	7.67	9.25	11.08
0.30	5.84	6.13	6.71	7.70	9.28	11.13
0.40	5.87	6.15	6.74	7.73	9.32	11.17
0.50	5.89	6.17	6.76	7.76	9.35	11.21
00101		e) 85 ⁰ C	1		10.0 m.3
0.02	5.59	5.86	6.41	7.42	8.97	10.73
0.05	5.60	5.87	6.42	7.43	8.99	10.74
0.07		0	6.43	7.44	9.00	10.75
0.10	5.61	5.88	6.44	7.45	9.01	10.77
0.15	5.63	5.90	6.46	7.47	9.03	10.79
0.20	5.65	5.91	6.47	7.49	9.05	10.82
0.30	5.67	5.94	6.51	7.52	9.09	10.87
0.40	5.70	5.97	5.54	7.56	9.13	10.92
0.50	5.73	6.00	6.57	7.59	9.17	10.96

 $\Delta S^* = (\Delta H^* - \Delta G^*)/T$

In Table 3 the values of ΔG^* for water-acetamide-KNO₃ ternary system are given only because the character of changes of ΔH^* and ΔS^* on concentration of KNO₃, contents of AcNH₂ and temperature is identical as in case of ΔG^* .

From literature [14-15] it follows that in case when the electrolyte orders the structure of water the free energy of activation of viscous flow increases with the rise of concentration of solution and inversely. As it can be seen from Table 3 in case of KNO₃ solutions in water-acetamide mixtures the values of ΔG^* at 25 and 40° C are practically constant. On the other hand the increase of acetamide contents in mixture causes the rise in value ΔG^* . One can be suppose that KNO₃ at 25 and 40° C does not change the interactions among water and acetamide molecules. The addition of acetamide causes the increase in ΔG^* . Probably it is connected with the formation of greater and greater amounts of mixed associates having threedimensional structure, which makes the viscous flow of solution more difficult. At 60° C and higher the values ΔG^* increase with the rise of concentration of KNO₃ in solution.

From this one can conclude that in this case KND_3 is positive solvated and it makes the viscous flow of solution more difficult. It is obvious that the increase of temperature of solution causes loosening of structure because of disordering it by stronger thermal motions of molecules. This effect makes easier the viscous flow of solution and the values of ΔG^* decrease.

The conclusions obtained in this paper are in agreement with the conclusion following from the analysis of B coefficient of the Jones-Dole's equation on concentration, composition of mixed solvent and temperature [3] for water-acetamide-KNO₃ system.

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60	Marian Woldan
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	Marian Woldan
	LEPKOŚĆ WZGLEDNA I ENERGIA SWOBODNA AKTYWACJI

LEPKIEGO PRZEPŁYWU ROZWTWORÓW KNO3 W MIESZANINACH WODA-ACETAMID W KILKU TEMPERATURACH

Przedyskutowano zależność lepkości względnej rozwtorów KNO₃ w mieszaninach woda-acetamid w zależności od stężenia roztworu,temperatury i składu mieszaniny. Obliczono i przedyskutowano zależność efektywnej molowej objętości lepkiego przepływu KNO₃ i energii swobodnej aktywacji lepkiego przepływu w zależności od stężenia, składu mieszaniny woda-acetamid i temperatury. Wysnuto wniosek o niszczącym wpływie KNO₃ na strukturę mieszanin woda-acetamid.