



Viscometric and Thermodynamics Study of Bi-Univalent Mixed Electrolytes in Aqueous Solutions at 293.15, 303.15 and 313.15 K

DURGESH GAUTAM*, VARSHA SHARMA and MUKHTAR SINGH

Department of Chemistry, Agra College, Agra-282 002, India

*Corresponding author: E-mail: dgautamchem@gmail.com

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Viscosities (η) and apparent molar volumes (V_ϕ) of the solutions of bi-univalent mixed electrolytes in aqueous solutions have been determined at 293.15, 303.15 and 313.15 K. From these data the values of coefficients A and B of Jones-Dole equation and that of V_ϕ^0 and S_ν of Masson's equation have been obtained. Besides the activation thermodynamic quantities $\Delta\mu_2^{0\#}$, $\Delta H_2^{0\#}$ and $\Delta S_2^{0\#}$ of viscous flow have also been calculated. The variation of ϕ_ν with y , fraction of ionic strength due to the first electrolyte in the mixture of two electrolytes at different temperatures is linear. From η and ϕ_ν data, the values of coefficients A and B of Jones-Dole equation and that of V_ϕ^0 and S_ν of Masson's equation have been obtained. Besides the activation thermodynamic quantities $\Delta\mu_2^{0\#}$, $\Delta H_2^{0\#}$ and $\Delta S_2^{0\#}$ of viscous flow have also been calculated for the mixed electrolytes in aqueous solutions. It is seen that the values of $\Delta\mu_2^{0\#}$ are much larger as compared to those of $\Delta\mu_1^{0\#}$ for all the bi-univalent mixed electrolytes. The values of $T\Delta S_2^{0\#}$ are negative while those of $\Delta H_2^{0\#}$ are positive and that $\Delta H_2^{0\#} > T\Delta S_2^{0\#}$. Thereby suggesting that ion-solvent interaction for the mixed electrolyte system, are nearly complete in the ground state. Further these the values of $T\Delta S_2^{0\#}$ and $\Delta H_2^{0\#}$ indicate the transition state is associated with the bond breaking and increase in order.

Keywords: Viscosities, Apparent molar volumes, Mixed electrolytes, Aqueous solution.

INTRODUCTION

The studies of thermodynamic properties of aqueous mixed electrolytic solutions using the Mayer-McMillan [1,2] theory as developed by Friedman and Anderson have been found to be useful in understanding the specific ion-ion interactions in solution. Patil and coworkers [3,4] have determined viscosities of aqueous mixed electrolytic solutions for the systems. KBr-NaBr, KBr-Bu₄NBr, NaCl-NaBr and NaCl-Bu₄NBr at various constant ionic strength with varying electrolyte mole fractions (y) at 25 °C.

It appears that studies on viscosities and apparent molar volume of bi-univalent mixed electrolytes in aqueous solution in relation to ion-ion and ion-solvent interaction are still lacking. With this aim, present study has been undertaken in aqueous solutions at different temperatures. The following bi-univalent mixed electrolytes have been used:

CaCl₂ + BaCl₂, NiCl₂ + BaCl₂, MgCl₂ + BaCl₂, MnCl₂ + CaCl₂, CaCl₂ + MgCl₂, NiCl₂ + MgCl₂, MnCl₂ + NiCl₂, NiCl₂ + CaCl₂, NiCl₂ + SrCl₂, Mg(NO₃)₂ + Ca(NO₃)₂, Zn(NO₃)₂ + Mg(NO₃)₂, Sr(NO₃)₂ + Co(NO₃)₂, Co(NO₃)₂ + Zn(NO₃)₂, BaCl₂ + MnCl₂, BaCl₂ + CoCl₂, NiCl₂ + CoCl₂, SrCl₂ + CoCl₂, CoCl₂ + MgCl₂ and SrCl₂ + MgCl₂.

The title study has been undertaken in the light of the following aspects:

(1) Determination of densities and viscosities of aqueous solutions of above mentioned bi-univalent mixed electrolytes at different temperatures (293.15, 303.15 and 313.15 ± 0.01 K) as a function of fraction of the ionic strength due to the first electrolyte in the mixture of two electrolytes (here in after denoted by y) keeping the ionic strength constant at a particular value depending upon the solubility of the electrolytes;

(2) Analysis viscosity data in the light of Jones-Dole equation;

(3) Determination of apparent molar volume (ϕ_ν) from the density data as a function of y , at different temperatures and calculation of the values of constants ϕ_ν^0 and S_ν of Masson's equation;

(4) Determination of free energies of activation for viscous flow $\Delta\mu_1^{0\#}$ and $\Delta\mu_2^{0\#}$ per mole of solvent and solute respectively and also the calculation of entropy of activation ($\Delta S_2^{0\#}$) and enthalpy of activation ($\Delta H_2^{0\#}$) of the bi-univalent mixed electrolytes solutions at different temperatures with a view to interpret the solution behaviour of mixed electrolytes; and

(5) Ascertaining structure-making or structure-breaking capacities of mixed electrolyte systems in aqueous solution.

EXPERIMENTAL

All the bi-univalent electrolytes were of analytical reagent grade and were used after drying in a vacuum oven at 110 °C for 10 to 12 h. The standard stock solutions of these electrolytes were prepared in doubly distilled water (specific conductivity: $10^{-6} \text{ ohm}^{-1} \text{ cm}^{-1}$). The solutions of mixed electrolytes of different composition with y varying in the range of 0.0 to 1.0, were prepared by mixing requisite volume of the solutions of individual electrolytes, in a measuring flask keeping the ionic strength constant.

The densities and viscosities of aqueous solutions of mixed electrolytes were determined at different temperatures (293.15, 303.15 and $313.15 \pm 0.01 \text{ K}$). The viscosity data were analyzed in the light of the modified form of Jones-Dole equation [5] as given below:

Jones-Dole equation is

$$\eta_{\text{rel}} = \frac{\eta}{\eta_0} = 1 + A\sqrt{C} + BC \quad (1)$$

$$\text{or} \quad \frac{(\eta_{\text{rel}} - 1)}{\sqrt{C}} = A + B\sqrt{C} \quad (2)$$

In the case of solutions of mixed electrolytes (at a constant ionic strength) replacing C by y *i.e.* fraction of ionic strength due to the first electrolyte in the mixtures of two electrolytes in eqn. 3, the Jones-Dole equation assumes the following modified form [6].

$$\frac{(\eta_{\text{rel}} - 1)}{\sqrt{y}} = A + B\sqrt{y} \quad (3)$$

Thus from the linear plots of $(\eta_{\text{rel}} - 1)/\sqrt{C}$ and $(\eta_{\text{rel}} - 1)/\sqrt{y}$ versus \sqrt{y} , the value of A and B can be obtained.

The apparent molar volume (ϕ_v) of aqueous solutions of bi-univalent mixed electrolyte systems was determined as a

function of y , at different temperatures using the following eqn. 4 [7]:

$$\phi_v = \frac{1000(\rho_0 - \rho)}{C\rho_0} + \frac{\bar{M}}{\rho_0} \quad (4)$$

where ρ_0 and ρ are the densities of solvent and solution respectively and C is the molarity of the solution, \bar{M} is the effective molecular weight of the mixed electrolyte system given by

$$\bar{M} = \frac{n_1 M_1 + n_2 M_2}{n_1 + n_2} \quad (5)$$

where n_1 and n_2 are the number of moles, M_1 and M_2 are the molecular weights of first and second electrolytes respectively. The variation of apparent molar volume (ϕ_v) with the molar concentration of the electrolyte solution is governed by Masson's equation (eqn. 6) [8]

$$\phi_v = \phi_v^0 + S_v \sqrt{C} \quad (6)$$

In the present study, the modified form of this equation in terms of y is to be determined on the basis of the linearity or otherwise of Φ_v versus \sqrt{y} plots Φ_v versus y plots.

Free energies of activities of viscous flow $\Delta\mu_2^{0\#}$ and $\Delta\mu_1^{0\#}$ per mole of solute and solvent respectively were determined at different temperatures. The values of entropy and enthalpy of activation of viscous flow were calculated from the following equations:

$$d \frac{(\Delta\mu_2^{0\#})}{dT} = -\Delta S_2^{0\#} \quad (7)$$

$$\Delta H_2^{0\#} = \Delta\mu_2^{0\#} + T\Delta S_2^{0\#} \quad (8)$$

The experimental data in regard to foregoing aspects of the present study have been presented in Tables 1-5.

RESULTS AND DISCUSSION

The densities and viscosities of aqueous solution of bi-univalent mixed electrolytes have been determined at 293.15,

TABLE-1
VALUES OF COEFFICIENTS A AND B OF JONES-DOLE EQUATION FOR BI-UNIVALENT MIXED ELECTROLYTE SYSTEMS IN AQUEOUS MEDIUM AT DIFFERENT TEMPERATURES

Mixed electrolyte system	Ionic strength (I)	A ($\text{dm}^{3/2} \text{ mol}^{-1/2}$)			B ($\text{dm}^3 \text{ mol}^{-1}$)		
		293.15 K	303.15 K	313.51 K	293.15 K	303.15 K	313.51 K
CaCl ₂ + BaCl ₂	3.0	-0.0675	-0.1551	-0.198	0.527	0.5294	0.5438
NiCl ₂ + BaCl ₂	3.0	-0.3461	-0.5698	-0.62	0.3801	0.5104	0.6332
MnCl ₂ + CaCl ₂	3.0	-0.0772	-0.1634	-0.2036	0.7668	0.7983	0.5357
BaCl ₂ + CoCl ₂	1.5	-0.3276	-0.4667	-0.5761	0.2848	0.3849	0.4809
NiCl ₂ + CoCl ₂	1.5	0.1451	0.0777	0.0646	0.1671	0.1875	0.1922
SrCl ₂ + CoCl ₂	1.5	-0.0711	-0.1354	-0.1491	0.5405	0.5548	0.5734
CoCl ₂ + MgCl ₂	1.5	-0.3809	-0.6266	-0.7107	0.4755	0.6366	0.7684
MgCl ₂ + BaCl ₂	3.0	0.1287	0.1063	0.0802	0.294	0.3065	0.3137
CaCl ₂ + MgCl ₂	3.0	-0.0282	-0.0417	-0.0542	0.1878	0.1894	0.2139
NiCl ₂ + MgCl ₂	3.0	-0.2799	-0.5497	-0.6049	0.3898	0.5823	0.7007
SrCl ₂ + MgCl ₂	1.5	-0.2984	-0.5531	-0.5763	0.405	0.6146	0.6879
Mg(NO ₃) ₂ + Ca(NO ₃) ₂	3.0	-0.038	-0.132	-0.2071	0.5242	0.5361	0.547
Zn(NO ₃) ₂ + Mg(NO ₃) ₂	3.0	0.3334	0.185	0.1599	0.369	0.3794	0.3875
Co(NO ₃) ₂ + Zn(NO ₃) ₂	3.0	-0.0164	-0.0642	-0.0856	0.2697	0.2766	0.2839
Sr(NO ₃) ₂ + Co(NO ₃) ₂	3.0	-0.1385	-0.1954	-0.3336	1.1214	1.1577	1.1746
MnCl ₂ + NiCl ₂	3.0	0.0296	0.0207	0.0189	0.0557	0.3081	0.3564
NiCl ₂ + SrCl ₂	3.0	-0.3783	-0.4859	-0.5420	0.423	0.4273	0.4898
NiCl ₂ + CaCl ₂	3.0	0.1188	0.0999	0.0929	-0.2636	-0.273	-0.2812
BaCl ₂ + MnCl ₂	1.5	-0.5566	-0.6147	-0.6593	0.3687	0.5885	0.6937

TABLE-2
VALUES OF LIMITING APPARENT MOLAR VOLUME (ϕ_v^0) AND EXPERIMENTAL SLOPE (S_v) OF
BI-UNIVALENT MIXED ELECTROLYTE SYSTEMS IN AQUEOUS MEDIUM AT DIFFERENT TEMPERATURES

Mixed electrolyte system	Ionic strength (I)	V_ϕ^0 (cm ³ mol ⁻¹)			S_v (cm ³ dm ^{3/2} mol ^{-3/2})		
		293.15 K	303.15 K	313.51 K	293.15 K	303.15 K	313.51 K
CaCl ₂ + BaCl ₂	3.0	110.81	113.05	116.79	-31.15	-32.695	-42.88
NiCl ₂ + BaCl ₂	3.0	128.43	130.81	132.33	-36.748	-40.5	-43.33
MnCl ₂ + CaCl ₂	3.0	87.088	88.89	90.375	-59.358	-80.91	-81.58
BaCl ₂ + CoCl ₂	1.5	219.41	224.08	229.42	-23.482	-33.077	-52.11
NiCl ₂ + CoCl ₂	1.5	214.67	218.53	220.62	23.549	21.275	15.023
SrCl ₂ + CoCl ₂	1.5	220.72	224.79	227.12	-24.454	-28.391	-39.06
CoCl ₂ + MgCl ₂	1.5	236.93	240.39	243.44	-33.567	-40.262	47.274
MgCl ₂ + BaCl ₂	3.0	110.92	114.4	117.3	59.157	57.256	50.694
CaCl ₂ + MgCl ₂	3.0	166.74	170.32	173.35	-87.48	-95.193	-102.2
NiCl ₂ + MgCl ₂	3.0	159.92	161.53	162.1	-46.45	-50.693	-55.06
SrCl ₂ + MgCl ₂	1.5	220.47	224.83	227.76	-26.247	-28.165	-36.47
Mg(NO ₃) ₂ + Ca(NO ₃) ₂	3.0	159.12	160.42	160.97	-18.469	-17.714	-15.47
Zn(NO ₃) ₂ + Mg(NO ₃) ₂	3.0	141.23	143.96	144.95	16.371	11.598	9.073
Co(NO ₃) ₂ + Zn(NO ₃) ₂	3.0	259.03	262.62	265.34	-7.234	-18.562	-19.51
Sr(NO ₃) ₂ + Co(NO ₃) ₂	3.0	143.63	144.13	144.33	-38.751	-34.558	-32.24
MnCl ₂ + NiCl ₂	3.0	75.014	77.273	78.651	50.207	47.542	37.948
NiCl ₂ + SrCl ₂	3.0	122.83	123.75	124.800	-13.062	-12.441	-11.63
NiCl ₂ + CaCl ₂	3.0	67.931	69.299	70.764	43.282	38.129	30.679
BaCl ₂ + MnCl ₂	1.5	240.61	241.29	242.16	-47.48	-46.551	-45.51

303.15 and 313.15 \pm 0.01 K as a function of y , the fraction of ionic strength due to first electrolyte in the mixture of two electrolytes, keeping the ionic strength constant (Tables 1-5).

TABLE-3
VALUES OF $\Delta\mu_2^{0\#}$ FOR BI-UNIVALENT MIXED
ELECTROLYTE SYSTEMS IN AQUEOUS
MEDIUM AT DIFFERENT TEMPERATURES

Mixed electrolyte system	Ionic strength (I)	$\Delta\mu_2^{0\#}$ (kJ mol ⁻¹)		
		293.15 K	303.15 K	313.51 K
CaCl ₂ + BaCl ₂	3.0	83.790	87.066	92.222
NiCl ₂ + BaCl ₂	3.0	65.272	86.826	107.283
MnCl ₂ + CaCl ₂	3.0	112.996	121.677	130.323
BaCl ₂ + CoCl ₂	1.5	65.732	95.783	113.022
NiCl ₂ + CoCl ₂	1.5	49.183	54.106	56.669
SrCl ₂ + CoCl ₂	1.5	100.470	54.106	56.669
CoCl ₂ + MgCl ₂	1.5	93.876	119.847	142.675
MgCl ₂ + BaCl ₂	3.0	52.312	56.179	59.272
CaCl ₂ + MgCl ₂	3.0	45.503	47.650	53.022
NiCl ₂ + MgCl ₂	3.0	70.901	101.199	121.242
SrCl ₂ + MgCl ₂	1.5	82.122	114.527	128.828
Mg(NO ₃) ₂ + Ca(NO ₃) ₂	3.0	89.941	94.604	99.022
Zn(NO ₃) ₂ + Mg(NO ₃) ₂	3.0	66.546	70.463	73.832
Co(NO ₃) ₂ + Zn(NO ₃) ₂	3.0	69.047	72.674	76.242
Sr(NO ₃) ₂ + Co(NO ₃) ₂	3.0	168.605	179.032	186.746
MnCl ₂ + NiCl ₂	3.0	38.431	51.226	59.854
NiCl ₂ + SrCl ₂	3.0	71.478	74.103	77.075
NiCl ₂ + CaCl ₂	3.0	42.393	45.221	47.858
BaCl ₂ + MnCl ₂	1.5	79.938	113.183	131.727

TABLE-4
FREE ENERGY OF ACTIVATION, $\Delta\mu_1^{0\#}$ (kJ mol⁻¹)
FOR WATER AT DIFFERENT TEMPERATURE

293.15 K	303.15 K	313.15 K
20.58	20.71	20.85

The viscosity data have been analyzed in the light of modified form of Jones-Dole equation as under:

$$\frac{(\eta_{rel} - 1)}{\sqrt{y}} = A + B\sqrt{y} \quad (9)$$

The values of coefficients A and B have been obtained from the linear plots of $(\eta_{rel} - 1)/\sqrt{y}$ versus \sqrt{y} by the method of least squares and listed in Table-1, shows that the values of A are negative for all the mixed electrolyte systems except for the mixed systems: NiCl₂ + CoCl₂, MgCl₂ + BaCl₂, Zn(NO₃)₂ + Mg(NO₃)₂, MnCl₂ + NiCl₂ and NiCl₂ + CaCl₂, for which the values are positive. From this it follows that ion-ion interaction are weak in aqueous solution of all the mixed bi-univalent electrolytes except for the mixed systems: NiCl₂ + CoCl₂, MgCl₂ + BaCl₂, Zn(NO₃)₂ + Mg(NO₃)₂, MnCl₂ + NiCl₂ and NiCl₂ + CaCl₂, for which the ion-ion interaction are strong.

The values of coefficients B are, however positive for the entire mixed bi-univalent electrolyte, which indicate the presence of strong ion-solvent interaction. The data of apparent molar volume (Φ_v) as a function of y , have been analyzed in the light of the modified form of Masson's equation:

$$\phi_v = \phi_v^0 + S_v y \quad (10)$$

From the linear plots of Φ_v versus y , the values of Φ_v^0 and S_v have been obtained by the method of least squares and presented in the Table-2. It is seen that the values of Φ_v^0 are positive and large. The positive values of Φ_v^0 indicate the presence of strong ion-solvent interaction [7]. It may be attributed to increased solvation of ions in the mixed electrolyte systems. Further with the rise of temperature the values of Φ_v^0 tend to increase, which shows the ion-solvent interaction are rendered increasingly stronger at elevated temperatures. This may be ascribed to the increased solvation of ions with the rise of temperature.

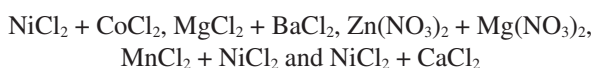
A perusal of Table-2 shows that the values of S_v are negative for the following mixed bi-univalent electrolyte:

TABLE-5
VALUES OF ENTROPY OF ACTIVATION ($\Delta S_2^{0\#}$) AND ENTHALPY OF ACTIVATION ($\Delta H_2^{0\#}$) FOR THE AQUEOUS SOLUTIONS OF BI-UNIVALENT MIXED ELECTROLYTE SYSTEMS AT DIFFERENT TEMPERATURES

Mixed electrolyte system	Ionic strength (I)	$\Delta S_2^{0\#}$ (kJ mol ⁻¹)			$\Delta H_2^{0\#}$ (kJ mol ⁻¹)		
		293.15 K	303.15 K	313.51 K	293.15 K	303.15 K	313.51 K
CaCl ₂ + BaCl ₂	3.0	-123.59	-127.81	-132.03	207.38	214.88	224.25
NiCl ₂ + BaCl ₂	3.0	-615.77	-636.38	-657.79	681.05	723.61	765.07
MnCl ₂ + CaCl ₂	3.0	-253.99	-262.65	-271.31	366.98	384.33	401.64
BaCl ₂ + CoCl ₂	1.5	-693.14	-716.79	-740.43	758.87	812.57	853.45
NiCl ₂ + CoCl ₂	1.5	-109.72	-113.47	-117.21	158.91	167.57	173.88
SrCl ₂ + CoCl ₂	1.5	-173.32	-179.24	-185.15	273.80	285.42	297.42
CoCl ₂ + MgCl ₂	1.5	-715.27	-739.67	-764.07	809.15	859.42	906.15
MgCl ₂ + BaCl ₂	3.0	-102.02	-105.50	-108.98	154.33	161.67	168.25
CaCl ₂ + MgCl ₂	3.0	-110.21	-113.97	-117.73	155.72	161.62	170.75
NiCl ₂ + MgCl ₂	3.0	-737.28	-763.05	-788.22	808.78	864.25	909.46
SrCl ₂ + MgCl ₂	1.5	-684.60	-707.95	-731.31	766.32	822.48	860.13
Mg(NO ₃) ₂ + Ca(NO ₃) ₂	3.0	-133.10	-137.64	-142.18	223.04	232.25	241.20
Zn(NO ₃) ₂ + Mg(NO ₃) ₂	3.0	-106.79	-110.43	-114.08	173.34	180.90	187.91
Co(NO ₃) ₂ + Zn(NO ₃) ₂	3.0	-105.46	-109.06	-112.66	174.51	181.73	188.90
Sr(NO ₃) ₂ + Co(NO ₃) ₂	3.0	-265.91	-274.91	-284.05	434.51	454.01	470.79
MnCl ₂ + NiCl ₂	3.0	-314.01	-324.72	-335.43	352.44	375.95	395.29
NiCl ₂ + SrCl ₂	3.0	-82.04	-84.84	-87.64	153.52	158.94	164.71
NiCl ₂ + CaCl ₂	3.0	-80.10	-82.83	-85.56	122.49	128.05	133.42
BaCl ₂ + MnCl ₂	1.5	-759.11	-785.00	-810.90	839.04	898.19	942.62

CaCl₂ + BaCl₂, NiCl₂ + BaCl₂, MgCl₂ + BaCl₂, MnCl₂ + CaCl₂, CaCl₂ + MgCl₂, NiCl₂ + MgCl₂, MnCl₂ + NiCl₂, NiCl₂ + CaCl₂, NiCl₂ + SrCl₂, Mg(NO₃)₂ + Ca(NO₃)₂, Zn(NO₃)₂ + Mg(NO₃)₂, Sr(NO₃)₂ + Co(NO₃)₂, Co(NO₃)₂ + Zn(NO₃)₂, BaCl₂ + MnCl₂, BaCl₂ + CoCl₂, NiCl₂ + CoCl₂, SrCl₂ + CoCl₂, CoCl₂ + MgCl₂ and SrCl₂ + MgCl₂.

The negative values of S_v for the above system indicate the presence of weak ion-ion interactions in the aqueous solution of these mixed bi-univalent electrolytes. On the other hand the positive values of S_v suggest the presence of strong ion-ion interaction in the aqueous solution of the following mixed bi-univalent electrolytes:



Free energies of activation for viscous flow: The free energies of activation of viscous flow, $\Delta\mu_1^{0\#}$ and $\Delta\mu_2^{0\#}$ per mole of solvent (water) and solute (mixed bi-univalent electrolytes) respectively are presented in the Tables 3A and 3B, respectively. It is seen that the values of $\Delta\mu_2^{0\#}$ for the aqueous solutions of all the bi-univalent mixed electrolyte are positive. Further the values of $\Delta\mu_2^{0\#}$ are much larger as compared to those of $\Delta\mu_1^{0\#}$ and thus $(\Delta\mu_2^{0\#} - \Delta\mu_1^{0\#}) > 0$. From this it follows that in aqueous solution all the mixed electrolytes under discussion are structure makers [9-11].

The entropy and enthalpy of activation of viscous flow for the aqueous solution of mixed bi-univalent electrolytes have been calculated and the values have been listed in Table-4. It is seen that in each case the values of $\Delta S_2^{0\#}$ are negative and those of $\Delta H_2^{0\#}$ are positive and thus $\Delta H_2^{0\#} > T\Delta S_2^{0\#}$. This suggests that ion-solvent interactions for the mixed electrolyte systems, under discussion are nearly complete in the ground state. Further the positive values of $\Delta H_2^{0\#}$ and negative values

of $\Delta S_2^{0\#}$ indicate that the transition state is associated with bond breaking and increase in order [12].

Conclusion

From the volumetric and viscometric studies reported in this paper, it has been concluded that the identical results are obtained. From both these studies with regard to ion-solvent and ion-ion interaction in aqueous solution of mixed bi-univalent electrolyte.

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