Thermodynamic function of valineacid in n-n dimethyl formamide mixtures from conductance measurement

*Sanaa T.Sarhan**

Abstract:

Some physical properties enthalpy (ΔH), entropy (Δs), free energy (ΔG),capacities(Δcp_o) and P_{ka} values) for valine in dimethyl foramideover the temperature range 293.15-318.15K, were determined by direct conductance measurements. The acid dissociation at six temperature was examined at solvent composition x_2) involving 0.141 of dimethyl foramide . As results, calculated values have been used to determine the dissociation constant and the associated thermodynamic function for the valine in the solvent mixture over temperatures in the range 293.15-318.15 k. The P_{ka1}, and P_{ka2} were increased with increasing temperature.

Key words: valine, Dimethyl formaide, Thermodynamic function, conductance measurement

Introduction:

Amino acid play many important roles in the living system. The interactions between the solvent and the various constituent group of a protein, play a cerucial role in the structure and function of proteins in aqueous solutions. Thermodynamic properties of amino acids in aqueous electrolyte solutions can provide valuable information regarding the confirmation stability of proteins in the solutions. The study of the behavior of the amino acids in aqueous solutions is a useful model for understanding the thermodynamic behavior of protein[1] [2]. In this paper, we study the dissociations constant and the associated thermodynamic properties of acid mean of investigation the change in the solute - Solvent interaction patterns that are attributed to the variation of the solvent composition [3]. Such investigation include valine and other amino acids in aqueous 1-Ppropyl-3- Methylim-idizolium Bromide Iconic liquid so solution of amino acids have also been reported as a function of temperature in the geochemical literature including densities ,molar , volumes ,viscosities, molar heat capacities and refraction indices. amino acid dissociation constant have also been reported as a function of temperature[4] [5]. In this paper, results are given for dissociation of valine in dimethyl formamide mixture at various temperature from direct conductance measurements.

Materials and Methods:
Materials:

Reagents used in this work were dimethyl formamide (DMF) of purity >99% obtained from Fluka was passed through a freshly prepared activated molecular sieve without any further purification .The solvent containing 40% w/w (DMF) was prepared by mass ratio using doubly distilled deionized water and then used for preparation of valine in required concentration .

*College of Science for Women, Baghdad University

Valine of purity 99% from (BDH), was used after re-crystallation from doubly distilled deionized water with a conductivity of approximately $0.7\mu S.cm^{-1}$ was used for preparation of solution[6].

Apparatus and Procedure:-

The samples were prepared by filling glass volumetric flask with composition of amino acid and (DMF) and then closing them tightly. The mass of chemicals used to prepare the mixture was determined with an uncertainty $1x\ 10^{-6}$ kg using Sertrious analytical balance AGTE2145. Theuncertainty in molalities of prepared solutions is approximately = $1x10^{-4}mol$ .kg^{-1} viscosity were determined using a suspended level Ubbelohde viscometer. the flow times were recorded electronically within electronic timer of precision $\pm$ 0.015 and the temperature of both was controlled to be better than $\pm$ 0.018 . the viscometer was calibrated with doubly distilled deionized water . flow times were reproducible to 0.01S[7]. Densities were determined using a vibrating-tube dosimeter Anton Paer (DMF 60/602) digital dosimeter, thermos total to +0.01 C^{o} the overall precision of measurements was estimated to be better then $\pm$ $2x10^{-6}$ g/ml .Tacussel electronic conduct meter , type CD810 , was used to measure the conductivity of the deionized water and of the valine. prepared solutions with an accuracy of $1\pm$ 10^{-9} $S.cm^{-1}$ The dielectric constant of the mixtures were determined using Radelkis precision dielectometer type OH-302 of maximum error on the dielectric constant scale of 2%. .densities of mixed solvents were measured with help of digital precision system DMA 60/602 (Anton Paer) the reproduce-bility of density measurement was $\pm$ $3x10^{-5}g.cm^{-3}$[8]. the density and the dielectric constant data of (DMF) at various temperatures have been used to estimate the appropriate values of the Debye-Huckel-Onsuger constant A and B using the relationships :-
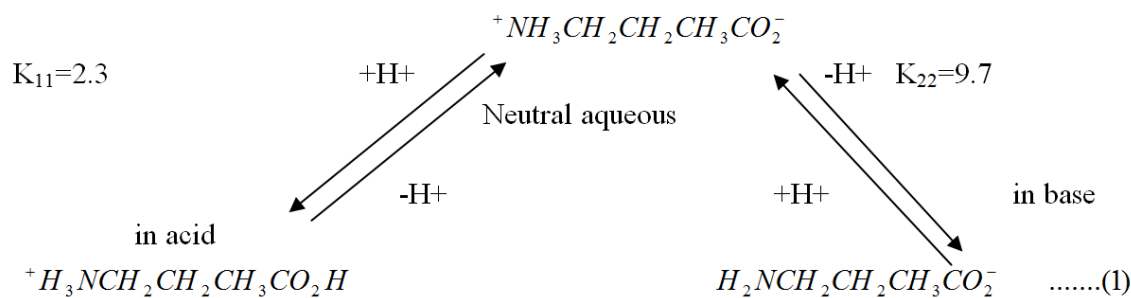
$$A = \frac{82.4}{\eta(DT)^{1/2}} \quad and \quad B = \frac{8.2 \ x10^5}{(DT)^{3/2}}$$

Where D, η and T represent respectively the dielectric constant, the viscosity and the temperatures , the resulting values of the constant A and B are presented in table (1) the solvent composition was expressed in terms of the mole fraction (x) of dimethyl formamide in the (DMF) +water mixture. The investigation covered on solvent compositions (x) which was 0.141 corresponding to 40% weight percentages of (DMF) in the mixture.

Results:
1-The degree of dissociation:
The acid dissociation of amino acids offer interesting examples of the relations between the function Ka and the true or 'thermodynamic' constant Ka of valine . In the dissociation of valine[9].

$$^{+}NH_3CH_2CH_2CH_3CO_2^{-}$$

$K_{11}=2.3$ +H+   Neutral aqueous   -H+ $K_{22}=9.7$

-H+   in acid   +H+   in base

$$^{+}H_3NCH_2CH_2CH_3CO_2H \qquad\qquad H_2NCH_2CH_2CH_3CO_2^{-} \quad ....(1)$$

The microscopic constants are K_{11}, K_{22} and so we define the first acid dissociation constant K_1, a macroscopic dissociation constant by

$$K_1 = \frac{(H^{+})[(^{+}H_3NCH_2CH_2CH_3CO_2^{-})+(H_2NCH_2CH_2CH_3CO_2H)]}{(^{+}H_3NCH_2CH_2CH_3CO_2H)} \qquad ...(2)$$

And second acid-dissociation constant is given by

$$K_2 = \frac{(H^{+})(H_2NCH_2CH_2CH_3CO_2^{-})}{[^{+}H_3NCH_2CH_2CH_3CO_2H+(H_2NCH_2CH_2CH_3CO_2^{-})]} \qquad ...(3)$$

An amino acid is said to be isoelectric at the pH at which there are equal concentrations of the positively and negatively charged forms setting ($H_3N^{+}CH_2COO^{-}$) and substituting from equations (2) and (3) yields

$$(H^{+})^2_{isoelectric} = K_1 K_2$$

Which may be written

$$pH_I = \frac{1}{2}(pK_1 + pK_2) \quad ...(4)$$

The onsager equation for incompletely dissociated electrolytes , can be written as [9,10].

$$\Lambda = \alpha[\Lambda_o - (A+B\Lambda_o)(\alpha C)^{1/2}] \ ...(5)$$

Where A and B are onsager constants and Λ_0 is the molar conductivity of the weak electrolyte at infinite dilution, Eqn.(5) may be written as:

$$\Lambda = \alpha \Lambda^{-} \quad ...(6)$$

Where

$$\Lambda^{-} = \Lambda_o - (A+B\Lambda_o)(\alpha C)^{1/2} \quad ...(7)$$

$$\Lambda^{-} = \Lambda_o - K(\Lambda c / \Lambda^{-})^{1/2} \ ...(8)$$

Λ^{-} is the molar conductivity of 1 mole of free ions, at the concentration C mole per liter, at the actual ionic concentration in the solution, where K , representing $A+B\Lambda_o$, is constant for a given solute in a particular solvent at a definite temperature. In order to estimate the value of α in Eqn.(6), an approximate value of Λ_o for amino acid, for a given solvent composition and temperature, was first made by extrapolating the experimental data of Λ^{-} against $(C)^{1/2}$. As a first approximation, Λ^{-} in the term. $(\Lambda c / \Lambda^{-})^{1/2}$ of Eqn.(8) was then taken as equal to Λ_o and hence a preliminary value of Λ could be derived from Eqn.(8), by utilizing the experimental value of Λ at the concentration (c). The result for Λ^{-} thereafter, inserted under the square root sign in Eqn.(8), thus obtaining a

better value of Λ^- in here of α. Using such values of Λ^- and α in Eqn.(7), it was possible to derive the correct values of Λ_o from the plots of Λ^- values against the corresponding values of (αc) ½ . Table (3) gives the resulting value of X_2 at a given temperature. The exact values of Λ_o could be derived, as mentioned earlier, from Λ^- , (αc)½ for solvent mixtures and temperatures and data obtained are presented in table(3). The values of Λ_o table(3)are then plotted against the experimental temperature (T) as indicated in Figure (1).Figure (1) shows that Λ_o at X_2=0.141 followed the sequence 308>303>298>293, implying an increase in Λ_o with increasing temperature.The change in Λ_o with change of temperature in the range 293–308 K is seen in Figure (1) to be almost linear suggesting a constant dependence of Λ_o values on temperature over the range referred to above . Thus, the increase of temperature invariably results in an increase of ion conductance. Since the conductance of an ion depends on its rate of movement, it seems reasonable to treat the conductance in a manner analogous to that employed for other processes taking at a definite rate which increases with temperature increasing , thus:

$$\Lambda_o = Ae^{-E/RT} \qquad ….(9)$$

Where A is constant, which may be taken as being independent of temperature over relatively small range; E is the activation energy of the processes which determines the rate of movement of ions, R is the gas constant and T is the temperature in Kelvin , Eq.no.(9) may be written as :

$$d \ln \Lambda_O /dt = (1/\Lambda_O)(d\Lambda_O /dt) = E/RT^2$$
and

$$E = \alpha RT^2 \quad ... (10)$$

Where α=(1/Λ_o)(dΛ_o/dt) is the temperature dependence of Λ_o. From the slopes of the lines in Figure (1) it was possible to evaluate the corresponding values of E, from Eqn.(10) , that cover the temperature range 293-318K, the such values of Λ_o have then been combined with the appropriate values of dΛ_o/dt to calculate the values of α and E . Table (4) gives the values $\alpha\ and\ E$ for Valineacid in Dimethyl form- amide at infinite dilutions.

2- The Dissociation Cconstant for theacid:
The relation between the function ka and the true or "thermodynamic" constant K of Valine may be written as :

$$K = Ka\ f+\ f- \ ...(11)$$

Where Ka = $\alpha^2 C / (1 - \alpha)$.
If the solution is sufficiently dilute for the Debye-Hukel law to applicable, it follows, for Valine type electrolyte, that

$$Log\ f^{\ +} = Log\ f^{-} = -A(\alpha C)^{1/2} \ ...(12)$$

The ionic strength, $1/_2 \sum c_i\ z_i^2$, being equal to

$$1/_2[(\propto cx_i^2) + (\propto cx_i^2)], i.e; to\ \alpha c.$$

Eqn.(12) may be expressed as :
$$Log\ K = Log\ Ka\ - 2A(\alpha C)^{1/2} \ .... (13)$$

The plot Log ka , obtained at various concentrations C, against (αC)$^{1/2}$ should be thus derive a straight line of intercept Log K .
Combining equations (6) and (13) , one may written :

$$Log\ K = log\left[\frac{\Lambda_2 c}{\Lambda-(\Lambda-\Lambda^-)}\right] - 2A(\Lambda c/\Lambda^-)^{\ 1/2} ... (14)$$

Since Λ^- for various concentration can be obtained from conductance data (table 2) and Onsager Eqn.(7), by method already described in the previous section, would be possible to derive the values of the dissociation function ka for variousacid concentration , and the ka_2 is obtained from the Eq.(13) :

$$Ka_2 = 4\alpha^2 C^2 / 1 - \alpha \qquad .... (15)$$

and, thereafter, calculatepI from Eqn.(4) . Such results for the dissociation of Valine in Dimethyl formamide mixture at different temperature are given in (table 5)

   The manner in which the $_pk$ values change with temperature as described in Figure (3) .Suggests a relatively large dependence of pk on temperature;

The dissociation constants (k_1 and k_2) of valine derived in the present work (table 5) are substantially less than those reported[11,12]. for theacid in water and in a number of other solvents. This is likely to be due to greater basicity and lower dielectric constant of DMF mixture than water.

The dependence of pk values for valine dissociation in DMF mixture an temperature could be expressed as:

$$_pK = \frac{A_1}{T} - A_2 + A_3 T \quad ... \quad (16)$$

Where A_1, A_2 and A_3 are constants for a given solvent composition (x=0.141) over the temperature range 293-308k Inserting the appropriate values of $_pk$ and temperature from table (5) in Eqn.(16), the values of the constant (A_1, A_2, A_3) could be calculated and the results are given in table (6).

3- Thermodynamic of theacid Dissociation: The standard thermodynamic function for valine dissociation in DMF mixture may be calculated from equation [13]:

$$\Delta G^O = (R \ln 10) (A_1 - A_2 T + A_3 T^2)... (17)$$

$$\Delta H^O = (R \ln 10) (A_1 - A_3 T^2) .... (18)$$

$$\Delta S^O = (R \ln 10) (A_2 - 2A_3 T) ... (19)$$

$$\Delta C_{PO}^o = (R \ln 10) (-2A_3 T) ... (20)$$

Table (7) summarizes the values of the standard thermodynamic functions which have been derived by Eq.(17-20) for the dissociation of the valine in DMF mixture at four temperatures From the following information could be obtained :

The value of ΔG^o is negative values for $_pk$ is refer to spontaneous reaction and high positive values of ΔG for $_pka$, at all temperature reflect the non spontaneous reaction .

The values of ΔH is high positive values at all temperature is reflect the large endothermic for $_pk_1$ and negative value for $_pk_2$ is reflect Exothermic step. The values of ΔS^o were generally positive which did not change significantly with the variation of temperature. $\Delta C^o p$ values were negative for solvents with $x_2=0.141$, $\Delta C^o p$ reflects, the effect of temperature on H^o so that one can write

$$C_P^o = dH^O / dT ...(21)$$

   It is shown in table(7) that the negative sign of ΔCp^o, is due to decreasing tendency of ΔH^o with the increase temperature .Thus the relatively more endothermic behavior lead to the greater increase in ΔS^o .

   The two sorts of interactions have to be considered; the first a maximum interaction between the two components of the solvent mixture and the second is the expected strong solute − solvent interaction due to the polarity and possibility of hydrogen bounds formation .

   The values of ΔS for $_pka$ decrease because the decomposition of

valine is very order the reactant at path second stage $_pka_2$.

Moreover, the high positive ΔH values at this stage indicates that the undissociatedacid molecules are highly stable so that the release of these molecules and subsequent dissociation stage should highly be endothermic.

Table(1):Dielectric constant(D) ,viscosity ,and the Onsager constants (A and B) for dimethyl formamide at six temperatures.

X	T/K	Dialectic	Viscosity	Values A	Values B
	293	63.01	2.338248	1.418005	0.3268874
	298	59.22	2.039288	1.264859	0.3497220
	303	60.32	1.810804	1.103171	0.3318375
0.1410	308	54.39	1.561893	0.994360	0.3781892
	313	62.056	1.408857	0.832971	0.3029156
	318	55.62	1.355498	0.839841	0.3485994

Table(2):Values of Λ (S mol^{-1}cm^2), Λ′(S mol^{-1}.cm^2),α and ka(mol.L^{-1}) for versus concentrations(mol L^{-1})of valine in dimethyl formamide + water mixture at different temperature(293-318)k at x_2=0.1410

T/K	Conc. mol.L^{-1}	ΛS mol^{-1}cm^2	Λ′S mol^{-1}.cm^2	A	Ka$_1$mol.L^{-1}	Ka$_2$ mol.L^{-1}	pPI
293	0.0183687	12.02	20.14184	0.596767723	0.016223098	0.00071134	0.008467219
	0.032785	8.601	20.01396	0.429750034	0.010617977	0.000598402	0.010917177
	0.0551302	5.754	19.95733	0.288315112	0.00643927	0.000409405	0.006643973
	0.08912	3.303	19.99606	0.165182541	0.002912809	0.000171519	0.002998568
	0.191326	1.5444	19.99429	0.077242053	0.001237069	7.3127605	0.001273632
298	0.0183687	11.198	18.85645	0.593855153	0.015949921	0.000695949	0.008322935
	0.032785	8.049	18.74356	0.429427494	0.010596051	0.000596718	0.01089441
	0.0551302	5.318	18.67903	0.284704291	0.006247294	0.000392225	0.006443407
	0.08912	3.067	18.71463	0.163882481	0.002862681	0.00016724	0.002946301
	0.191326	1.739	18.60999	0.093444435	0.001842835	0.000131787	0.001908729
303	0.0183687	11.269	19.20559	0.586756252	0.015303388	0.000659757	0.007981572
	0.032785	8.357	19.08497	0.437883843	0.011183222	0.000642186	0.011504316
	0.0551302	5.378	19.03644	0.282510806	0.006132596	0.000382058	0.006323624
	0.08912	3.171	19.05937	0.166374859	0.002959237	0.00017551	0.003046992
	0.191326	1.797	18.95996	0.094778681	0.001898631	0.000137716	0.001967489
308	0.0183687	13.23	21.79473	0.607027479	0.017223957	0.00076821	0.008996083
	0.032785	8.949	21.69511	0.412489266	0.009494775	0.000513609	0.00975158
	0.0551302	5.751	21.63724	0.265791755	0.005304608	0.000310917	0.005460067
	0.08912	3.682	21.61589	0.170337654	0.003116701	0.000189252	0.003211327
	0.191326	1.954	21.53393	0.09074052	0.001732562	0.000120316	0.00179272
313	0.0183687	14.645	22.14921	0.661197397	0.023702489	0.001151499	0.012426994
	0.032785	9.846	22.06828	0.446160734	0.011783495	0.000689447	0.012128219
	0.0551302	6.847	21.97954	0.311516984	0.007770694	0.000533815	0.008037601
	0.08912	6.468	21.73129	0.297635345	0.011240394	0.001192618	0.011836703
	0.191326	2.283	21.90009	0.104246147	0.002321161	0.000185182	0.002413752
318	0.0183687	17.007	27.45968	0.619344435	0.018510183	0.000842328	0.009676256
	0.032785	11.16	27.36626	0.407801431	0.009206728	0.000492367	0.009452912
	0.0551302	7.676	27.25641	0.281621828	0.00608652	0.000377994	0.006275517
	0.08912	4.892	27.23475	0.179623459	0.003505	0.000224433	0.003617217
	0.191326	2.648	27.12756	0.097612907	0.002020206	0.000150917	0.002095664

Table (3): Values of the molar conductivity dimethyl formamide at infinite dilution (Λ_o/S mo⁻l^1 cm^2) at x_2=0.141 and different temperature (293-318)K

Values of Λ_o						
X_2	293 K	298 K	303 K	308 K	313 K	318 K
0.141	22.041011	20.7402181	20.9784935	24.0464435	24.1064156	30.0099869

Table (4): The temperature coefficients (α) and the energies of activations (E) at infinite dilution as a function of the x_2 = 0.141

X_2	$d\Lambda_o$ / dt	Λ_o / S mol^{-1} cm^2	Slope	$E_{(kj,mole)}$	A
0.1410	-0.2451	0.568721963	-0.005288	0.10125374	1.304 x 10^{-7}

Table (5): Values of pk for the dissociation of valine in dimethyl formamaide + water mixture at six temperatures at x_2 =0.141

T/K	√αc	Pka$_1$	Pka$_2$	pPI
293	0.104698841	5.36489692	2.34300818	1.73576896
	0.118698588			
	0.12607486			
	0.121330409			
	0.121566496			
298	0.104443033	5.03714706	2.04243847	2.46144833
	0.118654037			
	0.125282898			
	0.120852003			
	0.133709947			
303	0.103816904	4.58962148	1.59375805	2.06472791
	0.119816617			
	0.124799348			
	0.121767514			
	0.134661152			
308	0.105595008	7.17103372	4.17627391	4.42734958
	0.116290415			
	0.121050207			
	0.123209138			
	0.131761226			
313	0.110205883	0.181801638	2.6973395	1.0620594
	0.120943705			
	0.13 1049585			
	0.162865779			
	0.141226762			
318	0.106660921	5.5778053	2.60359368	3.35330991
	0.115627721			
	0.124602840			
	0.126522894			
	0.136659749			

Table (6): Values of temperature dependence constants A_1, A_2 and A_3 at x_2= 0.141 of DMF+water.

Values of P	A_1	A_2	A_3
$_pka_1$	4.27×10^4	256	0.3955
$_pka_2$	3.99×10^4	241.9	0.3692
$_pPI$	4.58×10^4	302.7	0.4923

Table (7):Values of $_pK$ for the dissociation and the standard thermodynamics functions for the Valine in DMF+water mixture at six temperature.

| Values of P	T / K	ΔG^O kJ /	ΔH^O kJ /	ΔS^O J / mol	ΔCp^O kJ/K* mol
$_pka_1$	293	-31.1	-167	-464.0024166	4.436959693
	298	-29.0	-145	-388.2863808	4.512675728
	303	-2.72	-122	-312.5703451	4.588391764
	308	-25.9	-98.8	-236.8543094	4.6641078
	313	-24.9	-75.3	-161.1382737	4.739823835
	318	-48.8	-51.4	-85.42223801	4.815539871
$_pka_2$	293	-14.1	-157	-489.h16018	4.141910287
	298	-11.9	-137	-418.434956	4.21259135
	303	-99.6	-115	I-347.753893	4.283272413
	308	-84.0	-93.7	-277.07283	4.353953476
	313	-71.9	-71.8	-206.391767	4.424634539
	318	-29.5	-49.5	-135.710704	4.4953 15602
$_pPI$	293	-11.6	68.1	272.0838035	-5.522921
	298	-12.8	40.2	177.8360048	-5.6171688
	303	-13.4	11.9	83.5882061	-5.7114166
	308	-13.6	-16.9	-10.65959259	-5.8056644
	313	-13.3	-46.1	-104.9073913	-5.8999122
	318	116.4	-75.9	-199.15519	-5.99416
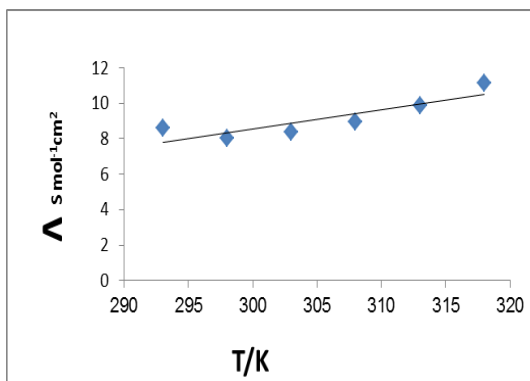
Fig (1) Molar conductivity (Smol^{-1}cm^2 of valine in DMF + water mixture as a function of temperature T
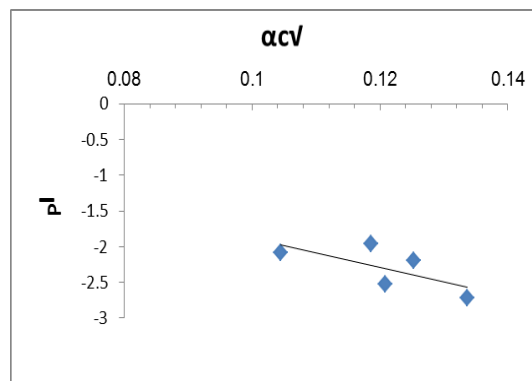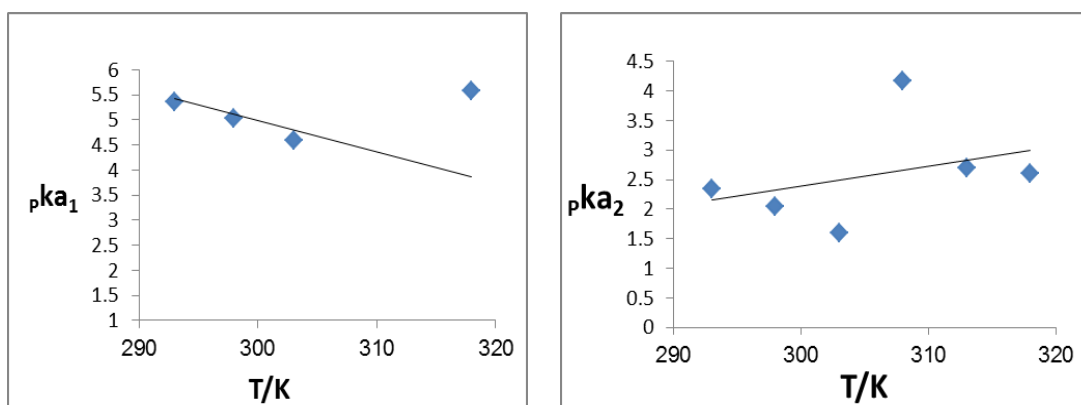

Fig.(2) log $_pI$versus$_l\alpha\sqrt{c}$, (C) expressed in moles per Liter

Fig(3) pK_{a1}, pK_{a2} and pH value of valine as a function of temperature T

Conclusion:

The done work showed that the dissociation constant (k_1 and k_2) of valine derived in present work are substantially less than those reported for theacid in water and a number of other solvents. The two sorts of interaction have to be considered; the first maximum interaction between the two components of the solvent mixture and the second is the expected strong solute –solvent interaction due to the polarity and possibility of hydrogen bounds formation .

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الدوال الحرارية لحامض الفاليين في مزيج الداي مثيل فورمامايد من قياسات التوصيلية

سناء طارق سرحان*

* كلية العلوم للبنات / قسم الكيمياء

الخلاصة :

تم تعيين بعض الخواص الفيزيائية مثل الانثالبي (ΔH)، والانتروبي (Δs)، والطاقة الحرة (ΔG) ، والسعة الحرة (Δcp_o) ، وثابت التفكك للحامض الفايلين pka في مذيب من داي مثيافورمامايد في مدى من درجات الحرارة K (15. 318.15-293)عن طريق قياسات التوصيلية مباشرة وعند كسر مولي للمذيب هو 0.141 من داي مثيل فورمامايد.ومن خلال النتائج لوحظ إن قيم ثابت التفكك لحامض الفاليين تزداد بزيادة درجة الحرارة.

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