THE AUDIO FREQUENCY CONDUCTANCE STUDY OF SOME METAL SUCCINATE SALTS IN AQUEOUS MEDIUM AT DIFFERENT TEMPERATURES (PART II: ZINC, NICKEL AND COBALT SUCCINATES)

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Summary

The electrical conductances of aqueous solutions of zinc, nickel and cobalt succinates have been measured at four temperatures in the temperature range from 298.15K to 313.15K. The limiting molar conductances (Λo), association constants (K_A) and the closest distances of approach (*a*) were calculated using the complete and modified forms of Fouss-Hsia (F/H) and Pitts (P). Quantitative results showed that these salts do not behave as "strong" electrolytes, and that their dissociations are far from complete. The abnormally law conductances of these electrolytes are due to the ion pair formation. The Walden product values, as well as the standard thermodynamic functions (ΔH^o, ΔG^o, ΔS^o) for association reactions have been evaluated.

Introduction

Following our investigations of association reactions of some succinate salts in aqueous medium **(Kosrat et.al. 2012**), we investigate the conductance behavior of Zinc, Nickel and Cobalt Succinates in the same medium at the temperature range from 298.15K to 313.15K. Information on ion-ion and ion-solvent interactions can be obtained from conductivity measurements. The limiting molar conductivity (Λo), association constants (K_A) and the closest distances of approach (***a***) were determined using the theoretical conductance equations of the complete and modified forms of Fouss-Hsia **(Fuoss et.al. 1959, Fuoss and HSIA 1967**) (F/H) and Pitts (P)**(FERNANDEZ 1969, PITTS 1953**). From the temperature depends of the association constant, thermodynamic quantities ΔH^o, ΔG^o and ΔS^o were evaluated. Also, Walden product ($\eta \Lambda_o$) was derived and discussed.

Experimental

Conductance measurements were made, as in part I **(Kosrat et.al. 2012**), at a range of temperatures between 298.15 - 313.15 K. All experimental techniques were described as before **(Kosrat et.al. 2012**). The cell constant (as determined by standard solutions of KCl) was 1.04079 cm^{-1}.

Zinc succinate was prepared by the same method as for copper succinate **(Kosrat et.al. 2012**). The product was anhydrated white crystalline powder. Elemental analysis confirmed the composition with experimental / calculated values (mass percent): C 27.3/26.5, H 2.3/2.2. The FT-IR spectra exhibits the following absorption bands (KBr pellet, υ/cm^{-1}), stretching frequencies corresponding to the carboxylate ion υ(CO_2^-) and υ(C-O) at 1695, 1552, 1418, 1310, cm^{-1}; the 1204 cm^{-1} absorption band is associated with υ(C-C), the following absorptions with metal-oxygen bonding and with υ(C-H) 918, 803, 715, 637, 583, 546, 438 cm^{-1}. The H-NMR δH = 2.30 ppm, the H-NMR was recorded in D_2O solvent.

The same procedure as for the preparation of both manganese and barium succinates was used again here **(Kosrat et.al. 2012**). The reaction was carried out between solutions of sodium succinate and nickel chloride. A light green tetra-hydrate crystal of nickel succinate was obtained as a final product. Elemental analysis indicated the composition with experimental / calculated values (mass percent): C 19.9/19.5, H 5.3/4.9. FT-IR spectra exhibits the following absorption bands (KBr pellet, υ/cm^{-1}); υ(O-H) bands at 3510, 3403 cm^{-1}; stretching frequencies corresponding to the carboxylate ion υ(CO_2^-) and υ(C-O) at 1547, 1462, 1407, 1331, 1287, 1245 cm^{-1}; the 1174 cm^{-1} absorption band is associated with υ(C-C) and following absorptions are with metal-oxygen bonding and with υ(C-H) 1036, 973,913, 806, 677 and 537 cm^{-1} **(Carine et.al. 2001**). The H-NMR δ_H = 2.40 ppm.

The same method, as for nickel succinate, was carried out here again by the reaction between cobalt chloride hexahydrate and a solution of sodium succinate. The reaction here was continued until the pH of the mixture become natural by carrying out evaporation. The

product of cobalt succinate as a tetra-hydrate was a light red crystalline powder. After filteration, it was washed by cold conductivity water until it become free from chloride ions, after that it was dried by air at room temperature. Elemental analysis confirmed the composition with experimental / calculated values (mass percent): C 20.0/19.5, H 5.4/4.9, FT-IR spectra exhibits the following absorption bands (KBr pellet, υ/cm^{-1}); υ(O-H) bands at 3519, 3411, cm^{-1}; stretching frequencies corresponding to the carboxylate ion $\upsilon(CO_2^-)$ and υ(C-O) at 1610, 1461, 1408,1328, 1286, 1243, cm^{-1}; the 1172 cm^{-1} absorption band is associated with υ(C-C) and the following absorptions with metal-oxygen bonding and with υ(C-H) 1036, 893, 802, 672 and 528[6] cm^{-1}.

All stock solutions were prepared by weight, and all measurements were done using the weight dilution technique. Figure (1) illustrates an example of FT-IR.

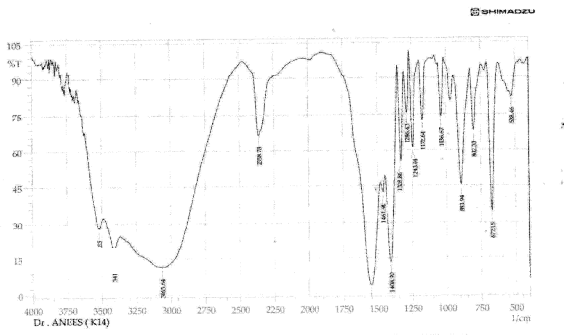

Figure (1) : FT-IR spectra for Co-Succinate

Results and Discussion

The measurement molar conductances (Λ) and the corresponding concentrations C in mol dm^{-3} at four different temperatures are given in tables 1-3. These data were analysed, as in part I **(Kosrat et.al. 2012**), using the complete and modified forms of both F/H and P equations.

Table (1): Experimental conductance data for Zn-succinate

298.15K		303.15 K		308.15		313.15 K	
Conc. * 10^3	**Λ**	**Conc. * 10^3**	**Λ**	**Conc. * 10^3**	**Λ**	**Conc. * 10^3**	**Λ**
0.1933	109.3060	0.5990	103.7540	0.2959	120.0800	0.5665	121.3440
0.4725	102.1620	0.6832	102.0040	0.7739	102.3920	0.7913	115.1500
0.5739	100.4340	0.7802	100.0000	0.8640	100.1580	0.8535	113.4620
0.7510	97.8740	0.8689	98.1760	0.9464	98.3060	0.9113	111.9800
0.8458	96.4420	1.0268	95.7520	1.0218	96.9140	0.9643	110.6780
0.9347	95.3420	1.0968	94.6220	1.0908	95.3080	1.0139	109.6220
1.0195	94.5600	1.1627	93.7280	1.1582	94.2600	1.0602	108.7680
1.0945	93.5480	1.2299	92.6300	1.2200	93.3260	1.1034	107.8080
1.2283	92.4660	1.2930	91.7340	1.2769	92.4260	1.1845	105.9220
1.2931	91.2560	1.3508	91.0840	1.3295	91.7060	1.2214	105.4860
1.3521	90.5400	1.4035	90.6300	1.3780	90.9280	1.2651	104.7200
1.4053	89.8940	1.4520	89.9300	1.4286	90.2600		
1.4550	89.6840	1.4993	89.3480	1.4780	89.7040		
1.5494	88.7520	1.5521	88.6560				

Λ ($ohm^{-1}cm^2\ mol^{-1}$); Conc. = concentration (mol dm^{-3})

Table (2): Experimental conductance data for Ni-succinate

298.15K		303.15 K		308.15		313.15 K	
Conc. * 10^3	**Λ**	**Conc. * 10^3**	**Λ**	**Conc. * 10^3**	**Λ**	**Conc. * 10^3**	**Λ**
0.2650	171.5260	0.8206	140.6000	0.8175	143.9180	0.4272	163.8480
0.3410	165.3260	1.0226	133.9500	0.8619	141.6360	0.6106	151.6960
0.5190	154.7500	1.1010	131.7360	0.9196	140.1020	0.6928	147.6040
0.6137	150.3780	1.1687	130.3420	0.9752	138.5180	0.7320	145.7460
0.7300	145.6600	1.2370	128.6140	1.0274	136.5440	0.7682	144.2840
0.8382	142.0840	1.3612	125.6720	1.1288	133.9640	0.8188	142.6800
1.0766	134.5460	1.4326	123.9460	1.1896	132.1460	0.8703	140.5240
1.2012	131.8460	1.5567	121.5900	1.2465	130.7080	0.9192	138.6960
1.5381	124.7920	1.6145	120.6200	1.2993	129.3940	0.9662	137.6120
1.6434	122.6560	1.7183	118.7820	1.3500	128.3940	1.0115	136.0800
1.8449	119.2740	1.7652	117.8380	1.4530	125.9180		
1.9398	118.1320			1.5049	124.8600		

Λ ($ohm^{-1}cm^2\ mol^{-1}$); Conc. = concentration ($mol\ dm^{-3}$)

Table (3): Experimental conductance data for Co-succinate

298.15K		303.15 K		308.15		313.15 K	
Conc. * 10^3	**Λ**	**Conc. * 10^3**	**Λ**	**Conc. * 10^3**	**Λ**	**Conc. * 10^3**	**Λ**
0.6062	155.4880	0.3984	169.4240	0.5756	157.3040	0.6351	155.0980
0.6693	151.7000	0.8004	147.0760	0.6878	152.4420	0.6894	152.6940
0.7292	150.2980	0.8447	145.8240	0.7401	150.1060	0.7382	150.3560
0.7844	148.0300	0.8901	143.9360	0.7912	148.2980	0.7838	148.5820
0.8364	146.2860	0.9352	142.8340	0.8398	146.8540	0.8648	145.4900
0.8846	144.7940	0.9719	141.7240	0.9187	143.2940	0.9045	144.2800
0.9332	142.8200	1.0087	140.6880	0.9535	142.9820	0.9424	142.9060
0.9772	141.9860	1.0423	139.6500	0.9864	141.9000	0.9781	141.9480
1.0186	140.3020	1.0764	138.8440	1.0168	140.7380	1.0111	140.9120
1.0579	139.5180	1.1008	137.8980	1.0690	139.2160	1.0386	140.1900
1.0941	138.7040						

Λ ($ohm^{-1}cm^2\ mol^{-1}$); Conc. = concentration ($mol\ dm^{-3}$)

The results of minimisation technique for the best values of the three parameters K_A, Λ_o and ***a*** are listed in table (4-6) together with the diffusion coefficient (D_{salt}), Walden product and thee corresponding values of the standard deviation (σ).

Table (4): Best fit results for Zn-succinate

Temps.	Parameter	F-H complete *	F-H modified *	Pitts complete	Pitts modified
298.15K	Λ_o	126.00	124.00	120.00	124.28
	K_A	70.0	90.0	300.0	60.0
	a	0.71	1.14	0.80	0.70

	$D_{salt} * 10^{-6}$	8.385	8.252	7.986	8.271
	□	0.2318	0.1923	0.2535	0.1804
	□ □$_o$	1.1225	1.1047	1.0691	1.1072
303.15K	□$_o$	140.00	141.00	134.00	136.00
	K_A	198.0	187.0	450.0	150.0
	a	0.69	0.71	1.10	0.89
	$D_{salt} * 10^{-6}$	9.473	9.541	9.067	9.202
	□	0.0774	0.2279	0.2382	0.2112
	□ □$_o$	1.1175	1.1255	1.0696	1.0856
308.15K	□$_o$	156.00	151.00	150.00	150.2
	K_A	392.0	270.0	700.0 and 705.0	260.0
	a	0.70	0.71	0.90	0.49
	$D_{salt} * 10^{-5}$	1.073	1.040	1.032	1.033
	□	0.0470	0.1008	0.2555	0.1438
	□ □$_o$	1.1276	1.0914	1.0842	1.0856
313.15K	□$_o$	176.00	168.00	162.00	156.00
	K_A	480.0	310.0	560.0	130.0
	a	1.01	1.19	1.40	1.28
	$D_{salt} * 10^{-5}$	1.230	1.174	1.132	1.090
	□	0.1438	0.1034	0.2804	0.1448
	□ □$_o$	1.1510	1.0987	1.0595	1.0202

* Preferred Values

Units of :

□$_o$ $ohm^{-1}cm^2 mol^{-1}$

K_A $dm^3 mol^{-1}$

a nm

D_{salt} $cm^2 s^{-1}$

□ unitless

□ □$_o$ $ohm^{-1} cm^2 mol^{-1}$ Cp

Table (5): Best fit results for Ni-succinate

Temps.	Parameter	F-H complete *	F-H modified *	Pitts complete	Pitts modified
298.15K	Λ_o	212.60	213.00	212.00and212.80	211.40
	K_A	370.0	420.0	780.0and790.0	410.0
	a	4.40	8.00	3.50	6.90
	$D_{salt} * 10^{-5}$	1.415	1.418	1.414	1.407
	β	0.0897	0.0894	0.2673	0.1919
	$\Lambda \Lambda_o$	1.8941	1.8976	1.8887 and 1.8958	1.8834
303.15K	Λ_o	217.00	216.00	215.60 and 215.80	217.20
	K_A	420.0	507.0	810.0	500.0
	a	0.40	1.10	1.00	0.50
	$D_{salt} * 10^{-5}$	1.468	1.462	1.460	1.470
	β	0.0827	0.0623	0.2249	0.1418
	$\Lambda \Lambda_o$	1.7321	1.7241	1.7209 and 1.7225	1.7337
308.15K	Λ_o	232.00	230.40	233.00	230.20
	K_A	595.0	600.0	1050.0	506.0
	a	0.60	1.39	0.30	0.41
	$D_{salt} * 10^{-5}$	1.596	1.585	1.602	1.583
	β	0.1041	0.1749	0.2526	0.2354
	$\Lambda \Lambda_o$	1.6769	1.6653	1.6841	1.6639
313.15K	Λ_o	242.00	240.20	240.00	240.40
	K_A	850.0	698.0	1190.0	681.0
	a	1.01	1.11	0.30	0.75
	$D_{salt} * 10^{-5}$	1.692	1.679	1.678	1.680
	β	0.1580	0.2768	0.2118	0.3727
	$\Lambda \Lambda_o$	1.5827	1.5709	1.5696	1.5722

* Preferred Values

Units of :

Λ_o $ohm^{-1}cm^2 mol^{-1}$

K_A $dm^3 mol^{-1}$

a nm

D_{salt} $cm^2 s^{-1}$

β unitless

$\Lambda \Lambda_o$ $ohm^{-1} cm^2 mol^{-1}$ Cp

Table (6): Best fit results for Co-succinate

Temps.	Parameter	F-H complete *	F-H modified *	Pitts complete	Pitts modified
298.15 K	□o	224.20	223.80	224.00	222.00
	K_A	560.0	505.0	850.0	460.0
	a	0.99	1.14	0.31	0.61
	$D_{salt} * 10^{-5}$	1.492	1.489	1.491	1.477
	□	0.1643	0.2263	0.2452	0.2365
	□ □o	1.9974	1.9938	1.9956	1.9778
303.15 K	□o	233.60	231.20	232.00	230.00
	K_A	600.0	550.0	890.0	530.0
	a	0.69	0.99	1.00	0.71
	$D_{salt} * 10^{-5}$	1.581	1.564	1.570	1.566
	□	0.1669	0.1845	0.2879	0.2186
	□ □o	1.8646	1.8454	1.8518	1.8359
308.15 K	□o	236.00	235.00	235.40	235.20
	K_A	610.0	579.0	1000.0	534.0
	a	0.71	1.19	0.30	0.55
	$D_{salt} * 10^{-5}$	1.623	1.616	1.619	1.618
	□	0.1835	0.3091	0.3218	0.3811
	□ □o	1.7058	1.6986	1.7015	1.7000
313.15 K	□o	242.80	240.00	240.20-241.20-242.20	239.00
	K_A	760.0	609.0	1060.0-1070.0-1090.0	545.0
	a	0.99	1.19	0.50	0.53
	$D_{salt} * 10^{-5}$	1.697	1.678	1.686	1.671
	□	0.0391	0.2429	0.3079	0.3030
	□ □o	1.5879	1.5696	1.5709- 1.5774 - 1.5840	1.5631

* Preferred Values

Units of :

□o $ohm^{-1}cm^2 mol^{-1}$

K_A $dm^3 mol^{-1}$

a nm

D_{salt} $cm^2 s^{-1}$

□ unitless

□ □o $ohm^{-1} cm^2 mol^{-1}$ Cp

All are identified in a part I**(Kosrat et.al. 2012**, **Oliver 2003)**. Standard thermodynamics quantities for the association reaction are obtained from the temperature dependence of the association constant K_A. The standard enthalpy change (ΔH^o) was determined from the slope of log K_A versus 1/T, while the standard Gibbs energy (ΔG^o) and entropy (ΔS^o) changes were calculated using the well known relations:- $-RT\ln K_A = \Delta G^o = \Delta H^o - T\Delta S^o$.

As well as in part I, it was noted that the minimisation Λ_o and ***a*** values for the complete and modified Fouss-Hisa equations were almost greater than those values obtained using the complete and modified forms of Pitts. The reason for this was mentioned.

A gain here, it was obvious from the minimisation technique that the parameter Λ_o had the greatest effect on the values of S^2 during the variation of the three parameters Λ_o, KA and ***a*** due to the fact that Λ_o is the leading term in all the conductance equations. As shown in table (5) and (6), each system gave a reasonable best set of parameters at each temperature. It is obvious that both Λ_o and K_A showed an expected trend with temperature, and this trend for ***a*** was quite irregular and covered a broader Range of values. The increase of Λ_o with an increase of temperature is due to the decrease of solvent viscosity, while the increase in K_A with increase in temperature is due to the decrease in dielectric constant which leads to the stabilization of ion-pair. The same trend was found by others **(Vesna s et.al.2005, Franchini et.al. 1987)**. With respect to the association constants given in tables(4-6), the same behaviors as for succinate salts mentioned in part I **(Kosrat et.al. 2012**), it can be anticipated that Zinc, Nickel and Cobalt succinates do not behave as "strong" electrolytes, and that their dissociation are far from complete. Furthermore, it can be generally predicated, for all three studied salts, that the degree of dissociation is changed from one to another.

Once more again, the three studies salts are more highly dissociated than oxalates and malonates due to the less degree of their donor property **(Kosrat et.al. 2012**).

In figures (2-4), curves of molar conductance versus square root of concentration at four temperatures have been obtained for the three succinate salts. For these electrolytes, the measured conductivity was abnormally small. Abnormally weak salts usually form auto-complexes readily, as is shown by Vosburgh and Beckman's solubility measurements **(Vosburgh et.al. 1940)** for Zinc and Cadmium oxalates and by Ives and Riley's conductivity measurements **(Ives et.al. 1931)** for copper malonate. In this part, with the exception of Zinc succinates, however, the experimental conductances give no evidence of this.

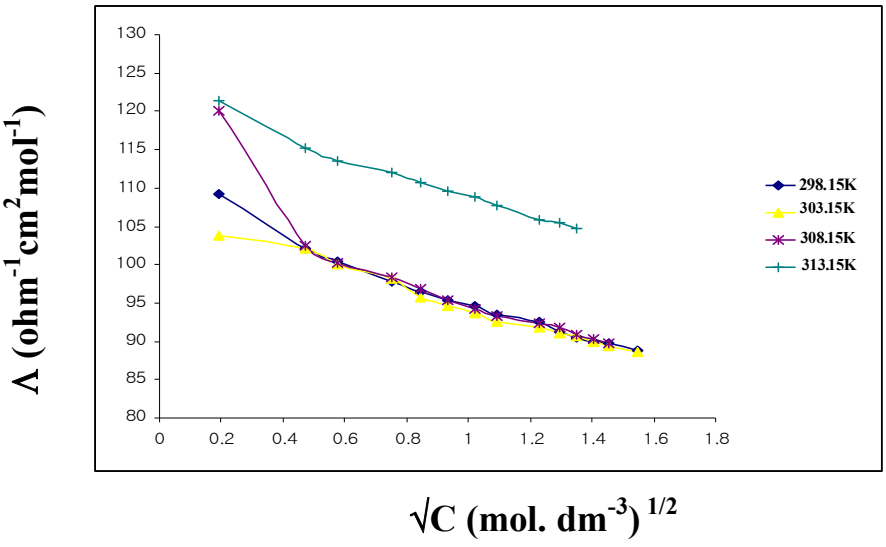

Figure (2): Molar conductance versus square root of concentration for Zn-succinate at different temperatures

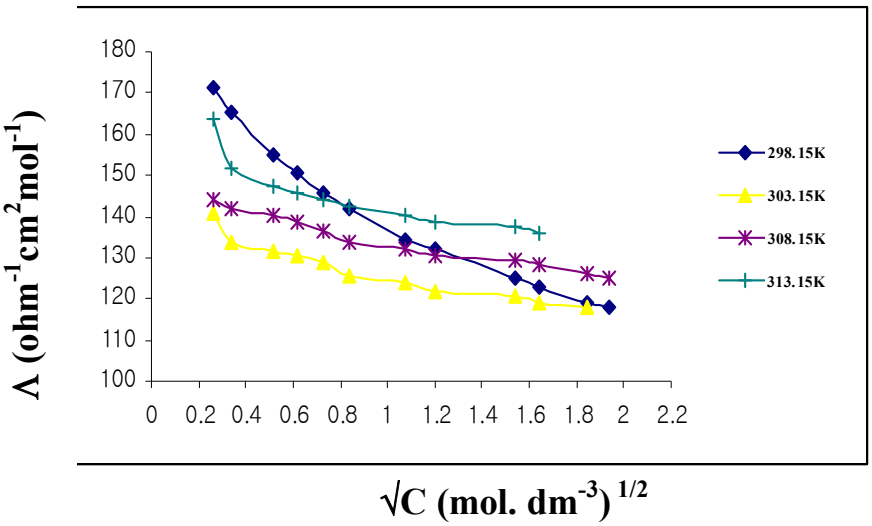

Figure (3): Molar conductance versus square root of concentration for Ni-succinate at different temperatures

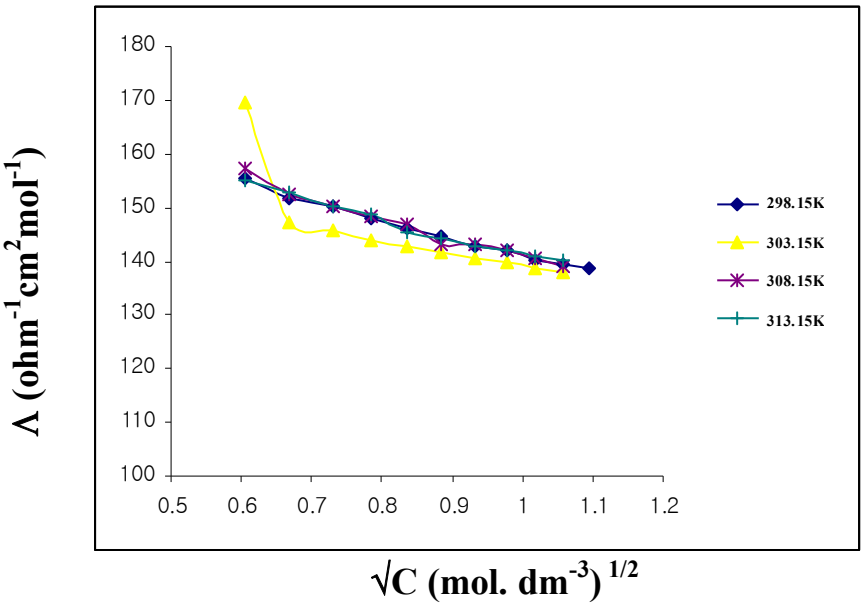

Figure (4): Molar conductance versus square root of concentration for Co-succinate at different temperatures

The same difficulties resulting from the limited solubilities, as in the case of the four succinate salts mentioned in part **(Kosrat et.al. 2012**), are a raised here also. Despite that, the values obtained for KA, Λ_o and ***a*** for both Nickel and Cobalt succinate are satisfactory, providing a particularly another good example of typical incompletely dissociated electrolytes the thermodynamic association constants of which conform to the law of mass action. The same conclusions **(Kosrat et.al. 2012**) for the abnormal low conductances of these electrolytes is predicated here as well.

As mentioned in part I **(Kosrat et.al. 2012**), no more recent determination of Λ_o at 298.15K for the studied succinate salts have been found. So, in comparison with ionic conductances at infinite dilution at 298.15K taken from reference **(Oliver 2003**):- $\lambda_{o1/2Zn}^{+2}$=52.80, $\lambda_{o1/2Ni}^{+2}$=49.60 and $\lambda_{o1/2Co}^{+2}$=55.00 together with the value of 58.8 **(Oliver 2003**) and 56.88**(Apelblat 2002)** of succinate ion, good agreement has been shown

for both Nickel and Cobalt succinates as shown in tables (5 and 6).

The Λ_o value obtained in this part for Zinc succinate is much lower than that given by direct combination of λ_o values of ionic conductances. In conclusion, it appears that the Zinc succinate is capable of showing either marked tendency to auto-complex formation according to the equation:-

$Zn^{+2}(C_4H_4O_4) + ZnC_4H_4O_4 \leftrightarrow [Zn(C_4H_4O_4)_2]^{2-} Zn^{2+}$

as is shown for zinc oxalates **(Vosburgh et.al.1940)**, or the zinc ions are linked by a bridging succinate ligand to form some kind of coordination polymer **(Zhao-Hui et.al. 2005)**.

Data from tables (4-6) show that the Walden product dependence on the temperature is substantially obeyed. The explanation is given in our part I.

The standard thermodynamics quantities for the association reaction of metal ion (M^{2+}) and succinate ions are obtained from the temperature dependence of K_A as given in tables (7-9), while figures (5-7) shown the corresponding plots for all three investigated succinate s

Table (7): Thermodynamic data for Zinc succinate

	ΔH^o (kJ mol^{-1})	ΔG^o (kJ mol^{-1})				ΔS^o (J $K^{-1}mol^{-1}$)			
		298.15 K	303.15 K	308.15 K	313.15 K	298.15K	303.15K	308.15K	313.15K
F-H Complete	17.23791	-10.531	-13.328	-15.298	-16.074	93.1374	100.8277	105.5847	106.3768
F-H Modified	15.21924	-11.154	-13.184	-14.343	-14.935	88.4563	93.6937	95.9346	96.2933
Pitts Complete	15.74554	-14.139	-15.398	-16.793	-16.475	100.2332	102.7331	105.5932	102.8917
Pitts Modified	12.15568	-10.149	-12.629	-14.246	-12.673	74.8103	81.7571	85.6780	79.2869

Table (8): Thermodynamic data for Nickel succinate

	ΔH^o (kJ mol^{-1})	ΔG^o (kJ mol^{-1})				ΔS^o (J $K^{-1}mol^{-1}$)			
		298.15K	303.15K	308.15K	313.15K	298.15K	303.15K	308.15K	313.15K
F-H Complete	7.059189	-14.658	-15.224	-16.367	-17.561	72.8398	73.5055	76.0220	78.6211
F-H Modified	6.585156	-14.973	-15.698	-16.389	-17.048	72.3064	73.5054	74.5551	75.4691
Pitts Complete	6.880383	-16.523	-16.879	-17.822	-18.437	78.4953	78.3750	80.1635	80.8475
Pitts Modified	6.881459	-14.913	-15.663	-15.952	-16.984	73.0990	74.3673	74.0985	76.2110

Table (9): Thermodynamic data for Cobalt succinate

	ΔH^o (kJ mol^{-1})	ΔG^o (kJ mol^{-1})				ΔS^o (J $K^{-1}mol^{-1}$)			
		298.15K	303.15K	308.15K	313.15K	298.15K	303.15K	308.15K	313.15K
F-H Complete	3.877289	-15.686	-16.123	-16.431	-17.270	65.6156	65.9749	65.9039	67.5309
F-H Modified	3.513957	-15.430	-15.903	-16.297	-16.693	63.5383	64.0507	64.2900	64.5280
Pitts Complete	3.67708	-16.720	-17.116	-17.697	-18.169	68.4121	68.5901	69.3626	69.7624
Pitts Modified	3.793577	-15.198	-15.810	-16.090	-16.404	63.6981	64.6663	64.5256	64.4981

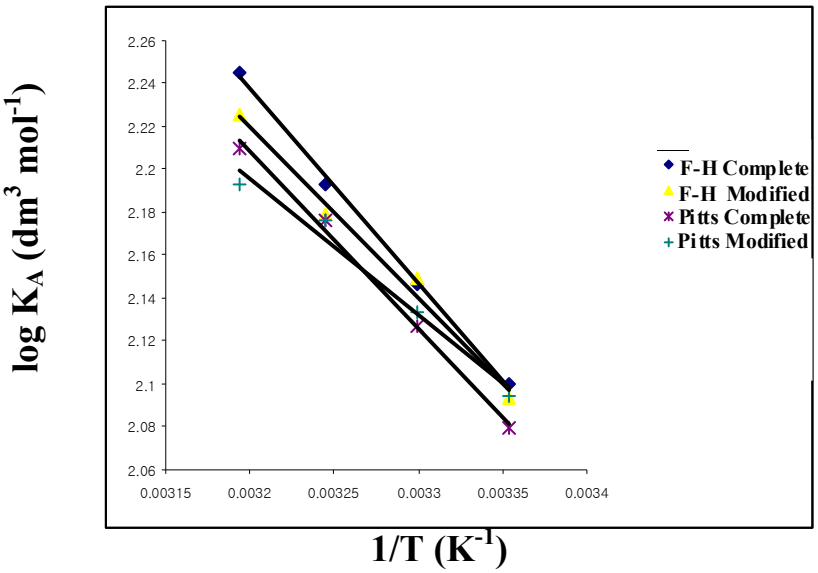

Figure (5): Logarithm of association constant versus inverse temperature for Zn-succinate

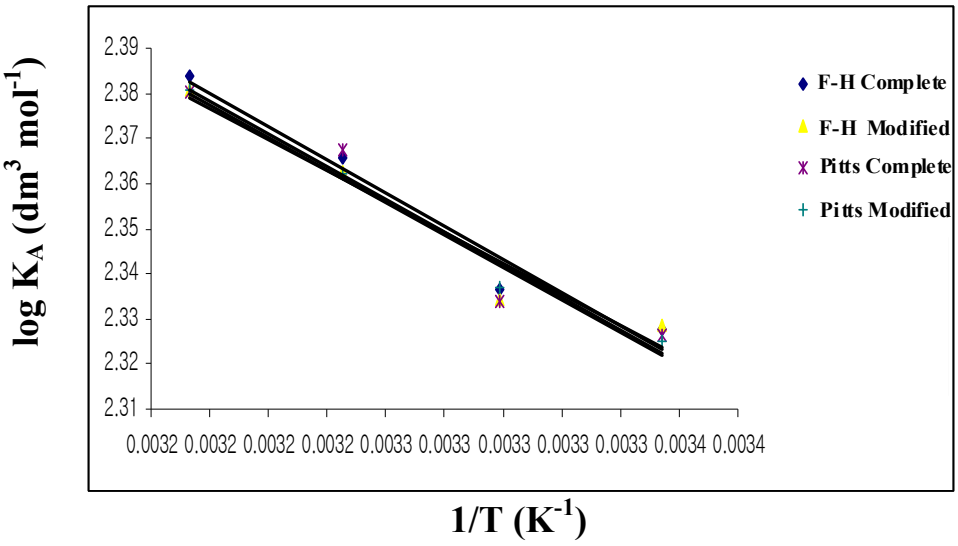

Figure (6): Logarithm of association constant versus inverse temperature for Ni-succinate

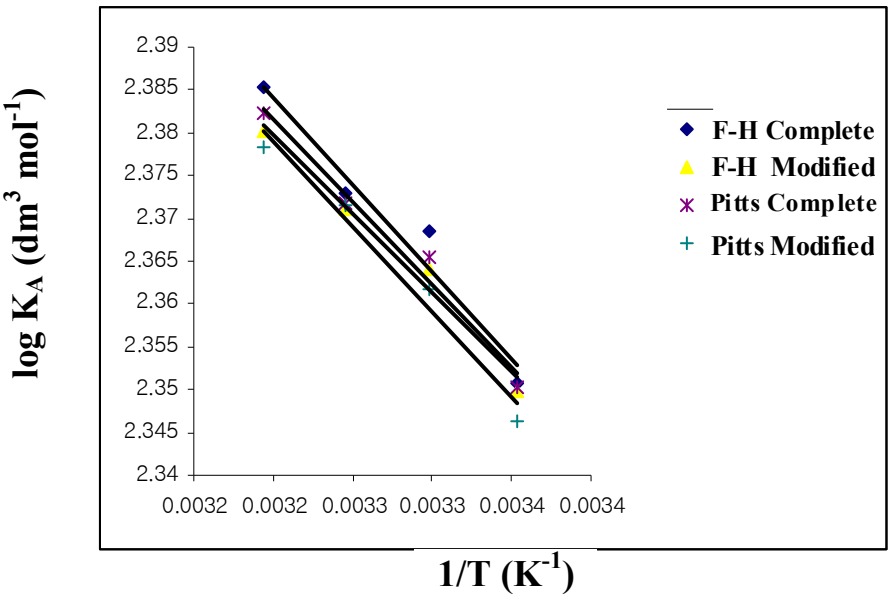

Figure (7): Logarithm of association constant versus inverse temperature for Co-succinate

The same interpretations and conclusions are recommended here as given in part I.

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