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Gbbs free energies for interaction of $CuCl_2$ with (E)-1-phenyl-2-(2-(4-((E)-pheyldiazenyl) phenyl hydrazono)-2-(phenylsulfonyl) ethanone in ethanol at different temperatures.

E.A.Gomaa, H.M.Abu El-Nader and Sh.E.Rashed

Chemistry Department, Faculty of Science, Mansoura University, 33516 Mansoura, Egypt.

------ABSTRACT-----

the association constants, formation constants and Gibbs free energies of solvation are calculated from the conductometric titration curves of $CuCl_2$ with (E)-1-phenyl-2-(2-(4-((E)- henyldiazenyl)phenyl)hydrazono)-2-(phenylsulfonyl)ethanone(L) in absolute ethanol at 293.15, 298.15, 303.15 and 308.15 K. On drawing the relation between molar conductance and the ratio of metal to ligand concentrations, different lines are obtained indicating the formation of 1:2, 1:1 and 2:1 (M:L) stoichiometric complexes. The formation constants and Gibbs free energies of these different complexes in absolute ethanol at 293.15, 298.15, 303.15 and 308.15 K follow the order: $K_f(2:1) > K_f(1:1) > K_f(1:2)$ for (M:L); and $\Delta G_f(2:1) > \Delta G_f(1:1) > \Delta G_f(1:2)$ for (M:L).

Key words: Association constant; formation constant; Gibbs free energy of solvation.

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I. INTRODUCTION

The long range ion - ion interactions due to screened columbic forces are the most important features of electrolyte in solutions. These act together with shorter - ranged forces between the solvent molecules and between the solvent molecules and ion. Electrical conductivity (EC) is a measure measure of solvent to conduct electric current and depends on: concentration of the ions, ligand and temperature in solutions. Current is carried out by both cations and anions, but to different degree. The conductivity due to divalent cations is more than that of mono-valent cations, it is not true for anions. Metal cations with do noble gas electron configuration (alkali and alkaline earth) metal ions together with the inert molecular ions like tetraalkylammonium,-phosphonium,-arsonium, and trialkylsulfonium ions exhibit properties mainly determined by their charge and size [1]. Solvation of such cations in protic and polar solvents is due essentially to electrostatic ion-dipole and ion induced dipole interactions. Metal cations with filled d-orbitals, the d 10 cations, exhibits partially covalent character in their interactions; their properties depend on the charge and size and partially ontheir electronegativity. Cations with incomplete d- orbitals called dⁿ-cations. With these cations protic and polar solvent molecules are strongly bound in complexes to a central cation through p-d orbital overlap and exchange only slowly with the bulk solvent. Therefore conductivity study is valuable on using transition metal cations [2.3].

The synthesis of transition metal complexes with Schiff bases of nitrogen, oxygen and sulfur donor has stimulated interest due to their vast variety of biological activities ranging from pharmacological, antitumor, fungicide, bactericide, anti-inflammatory, and antiviral activities [4,5]. Cobalt is the active center of coenzymes called cobalamins, the most common example of which is vitamin B12. As such it is an essential trace dietary mineral for all animals. Cobalt in inorganic form is also an active nutrient for bacteria, algae and fungi [6,7]. The aim of this work, consists in evaluation of the non-covalent behavior of CoCl2 in presents of (E)-1-phenyl-2-(2-(4-((E)-phenyldiazenyl)phenyl)hydrazono)-2-(phenylsulfonyl)ethanone (L1) in absolute ethanol solutions at 293.15, 298.15, 303.15 and 308.15 K. these non-covalent interactions can help us for analysis of salts role influence in bodies and environment. This work provide the analytical analyst and the biological analyst data can help him for determing the concentration of CoCl₂ in blood and different solutions.

Experimental

Material and methods

All manipulations were performed under aerobic conditions. The copper chloride and the used reagents were Merck pure.

The ligand

(E)-1-phenyl-2-(2-(4-((E)-phenyldiazenyl)phenyl)hydrazono)-2-(phenylsulfonyl)ethanone

Conductometric titration

The conductometric titration of the ligand $(1x10^{-3})$ mole/L against CuCl₂ $(1x10^{-4})$ mole/L in absolute ethanol was performed with 0.5 ml interval additions from CoCl₂ solution. The specific conductance values were recorded using conductivity bridge HANNA, H1 8819N with a cell constant equal to 1. The conductometer was conducted with a thermostat of the type the Kottermann 4130 ultrathermostat. The temperature was adjusted at 293.15, 298.15, 303.15 and 308.15 K.

Results and discussion

The specific conductance values (K_s) of different concentrations of $CoCl_2$ in absolute ethanol were measured experimentally in absence and in presence of ligand at 293.15, 298.15, 303.15 and 308.15 K.

The molar conductance ($\Lambda_{\rm m}$) values were calculated [8] using equation

$$\Lambda_{\rm m} = (K_{\rm s}-K_{\rm solv}) \text{ Kcell x } 1000/c (1)$$

Where K_s and K_{solv} is the specific conductance of solution and the solvent, respectively; K_{cell} is the cell constant and C is the molar concentration of the $CoCl_2$ solutions.

The limiting molar conductance (Λ_0) at infinite dilutions were estimated for CoCl₂ in absolute ethanol alone(Fig. 1) and in the presence of the ligand (L 1)(Fig. 2) by extrapolating the relation between (Λ_m) and $C_m^{1/2}$ to zero concentration. By drawing the relation between molar conductance (Λ_m) and the molar ratio of metal to ligand (M/L) concentrations (Fig. 3- 6), different lines are obtained with sharp breaks indicating the formation of 1:2, 1:1 and 2:1 (M:L) stoichiometric complexes. The experimental data of (Λ_m) and (Λ_0) were analyzed for the determination of association and formation constants for each type of the stoichiometric complexes.

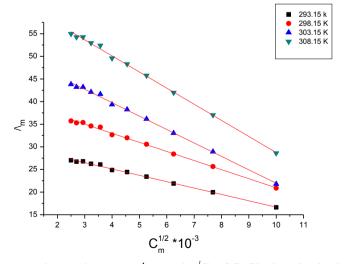


Fig. 1 The relation between molar conductance (Λ _m) and (\sqrt{C}) of CuCl₂ alone in absolute ethanol at 293.15, 298.15, 303.15 and 308.15 K.

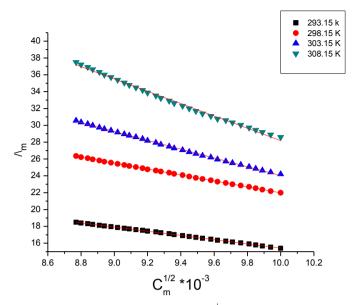


Fig. 2 The relation between molar conductance (Λ_m) and (\sqrt{C}) of CuCl₂ in the presence of ligand (L2) in absolute ethanol at 293.15, 298.15, 303.15 and 308.15 K.

The association constants of $CoCl_2$ in the presence of the ligand (L2) in absolute ethanol at 293.15, 298.15, 303.15 and 308.15 K for asymmetric electrolytes were calculated [9,10] by using equation: $K_A = [\Lambda_0^2 (\Lambda_0 - \Lambda_m)]/[4C_m^2 + \Lambda^3 S(z)]$ (2)

Where (Λ_m , Λ_0) are the molar and limiting molar conductance, respectively of $CoCl_2$; C_m is molar concentration of $CuCl_2$, S(Z) is Fuoss-Shedlovsky factor, equal with unity for strong electrolytes [11]. The calculated association constants are shown in Table 1.

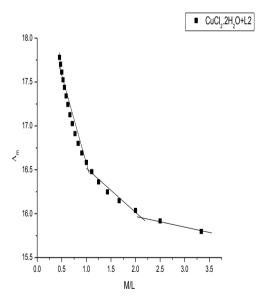


Fig. 3 The relation between molar conductance (Λ_m) and the molar ratio (M/L) of CuCl₂ in presence of (L2) in absolute ethanol in 293.15 K.

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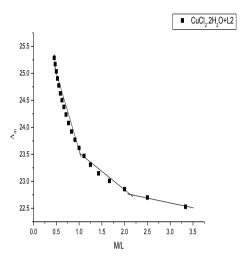


Fig. 4 The relation between molar conductance (Λ_{m}) and the molar ratio (M/L) of CuCl₂ in presence of (L2) in absolute ethanol in 298.15 K.

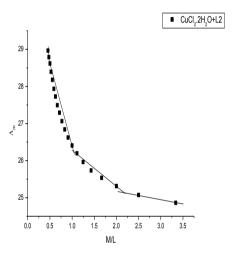


Fig. 5 The relation between molar conductance (Λ_m) and the molar ratio (M/L) of CuCl₂ in presence of (L2) in absolute ethanol in 303.15 K.

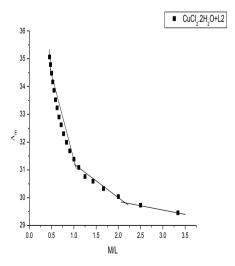


Fig. 6 The relation between molar conductance (Λ_{m}) and the molar ratio (M/L) of CuCl₂ in presence of (L2) in absolute ethanol in 308.15 K.

The Gibbs free energies of association (ΔG_A) were calculated from the associated constant [12,13] by applying equation :

$$\Delta G_{A} = -RT \ln K_{A} \tag{3}$$

Where R is the gas constant and T is the absolute temperatures (293.15, 298.15, 303.15 and 308.15 K). The calculated Gibbs free energies were presented in tables (1-4).

Table (1): Association constants of association of CuCl₂ with L2 at different temperatures.

$C_{[ligand]}$	C _{m[CuCl2]}	K _A				
	Temp	293.15 K	298.15 K	303.15 K	308.15 K	
0.99E-05	9.09E-05	3.22E+08	3.1E+08	6.14E+08	9.15E+08	
1.96E-05	8.33E-05	3.72E+08	3.58E+08	7.09E+08	1.05E+09	
2.91E-05	7.69E-05	4.24E+08	4.09E+08	8.09E+08	1.2E+09	
3.84E-05	7.14E-05	4.79E+08	4.62E+08	9.12E+08	1.35E+09	
4.76E-05	6.67E-05	5.34E+08	5.17E+08	1.01E+09	1.49E+09	
5.66E-05	6.25E-05	5.94E+08	5.75E+08	1.12E+09	1.65E+09	
6.54E-05	5.88E-05	6.56E+08	6.36E+08	1.23E+09	1.81E+09	
7.41E-05	5.56E-05	7.16E+08	6.94E+08	1.33E+09	1.98E+09	
8.26E-05	5.26E-05	7.79E+08	7.56E+08	1.44E+09	2.14E+09	
9.09E-05	0.00005	8.43E+08	8.17E+08	1.55E+09	2.29E+09	

Table (2): Gibbs free energies of association of CuCl₂ with L2 at different temperatures.

$C_{[ligand]}$	C _{m[CuCl2.2H2O]}	$\Delta G_A(k \text{ J/mol})$			
	Temp	293.15 K	298.15 K	303.15 K	308.15 K
0.99E-05	9.09E-05	-47.8997	-48.6198	-51.1688	-53.0362
1.96E-05	8.33E-05	-48.2517	-48.9827	-51.5316	-53.3987
2.91E-05	7.69E-05	-48.5741	-49.3152	-51.8640	-53.7311
3.84E-05	7.14E-05	-48.8713	-49.6180	-52.1663	-54.0328
4.76E-05	6.67E-05	-49.1388	-49.8965	-52.4282	-54.2927
5.66E-05	6.25E-05	-49.3970	-50.1601	-52.6819	-54.5450
6.54E-05	5.88E-05	-49.6415	-50.4096	-52.9228	-54.7850
7.41E-05	5.56E-05	-49.8534	-50.6260	-53.1285	-55.0242
8.26E-05	5.26E-05	-50.0618	-50.8390	-53.3307	-55.2172
9.09E-05	0.00005	-50.2531	-51.0344	-53.5165	-55.3951

The association free energies evaluated for CuCl₂ –ligand complexes are small and spontaneous indicating electrostatic attraction.

The formation constants (K_f) for $CoCl_2$ complexes were calculated for each type of complexes (1:2), (1:1) and (2:1) (K:L) by using equatin [14,15]:

$$K_f = [\Lambda_M - \Lambda_{obs}]/[(\Lambda_{obs} - \Lambda_{ML})[L]] (4)$$

Where Λ_m is the molar conductance of $CoCl_2$ alone, Λ_{obs} is the molar conductance of the solution during titration and Λ_{ML} is the molar conductance of the complex.

The obtained values (K_f) for $CuCL_2$ -ligand stoichiometric complexes are presented in tables 2,3. The Gibbs free energies of formation for each stoichiometric complexes were calculated by using the equation: $\Delta G_f = -RT \ln K_f$

The calculated ΔG_f values are presented in tables (5-16) [16-18].

Table.3 (a): Formation constants and Gibbs free energies of formation for $2:1\ (M/L)$ complexes in absolute ethanol at $293.15\ K$.

$(\text{cm}^2.\text{Ohm}^{-1})$	[L]	$(\wedge_{\text{obs}} - \wedge_{\text{ML}})$ [L]	$(\land_{M} - \land_{obs})$	$\mathbf{K_f}$	Δ G _f (k J/mol)
16.14746	5.66E-05	6.24E-06	14.5899	2337433	-35.8573
16.24727	6.54E-05	1.37E-05	14.49009	1054600	-33.9112
16.36161	7.41E-05	2.40E-05	14.37575	597986.4	-32.5239
16.47820	8.26E-05	3.64E-05	14.25916	391431.4	-31.4878
16.58377	9.09E-05	4.97E-05	14.15359	284866.3	-30.7107

 $\Lambda_{\rm ML} = 16.04 \, {\rm cm}^2.0 \, {\rm hm}^{-1}.$

Table 3(b): Formation constants and Gibbs free energies of formation for 2:1 (M/L) complexes in absolute ethanol at $298.15~\mathrm{K}$

/ _{obs} (cm ² .Ohm ⁻¹)	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	$\mathbf{K}_{\mathbf{f}}$	Δ G _f (k J/mol)
23.0076	5.66E-05	8.74E-06	18.0088	2060866	-36.1557
23.1473	6.54E-05	1.92E-05	17.8690	928966.7	-34.1741
23.3074	7.41E-05	3.37E-05	17.7090	526173.5	-32.7605
23.4706	8.26E-05	5.10E-05	17.5457	344042.6	-31.7039
23.6184	9.09E-05	6.96E-05	17.3979	250120.3	-30.9110

 $\Lambda_{\rm ML} = 22.85 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.3(c): Formation constants and Gibbs free energies of formation for 2:1 (M/L) complexes in absolute ethanol at 303.15K.

\wedge_{obs} (cm ² .Ohm ⁻¹)	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	$\mathbf{K}_{\mathbf{f}}$	Δ G _f (k J/mol)
25.53434	5.66E-05	1.25E-05	26.08277	2089347.0	-36.7967
25.73395	6.54E-05	2.75E-05	25.88316	941921.1	-34.7822
25.96264	7.41E-05	4.81E-05	25.65447	533573.0	-33.3452
26.19582	8.26E-05	7.29E-05	25.42129	348922.7	-32.2712
26.40696	9.09E-05	9.94E-05	25.21015	253699.7	-31.4653

 $\Lambda_{\rm ML} = 25.314 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.3 (d): Formation constants and Gibbs free energies of formation for 2:1 (M/L) complexes in absolute ethanol at 308.15K.

∧ _{obs} (cm².Ohm⁻¹)	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	$\mathbf{K}_{\mathbf{f}}$	Δ G _f (k J/mol)
30.32874	5.66E-05	1.62E-05	34.00085	2095151	-37.4108
30.58824	6.54E-05	3.57E-05	33.74135	944532.9	-35.3630
30.88553	7.41E-05	6.25E-05	33.44406	535069.9	-34.3213
31.18866	8.26E-05	9.47E-05	33.14093	349911.0	-33.0552
31.46315	9.09E-05	1.29E-04	32.86644	254422.2	-32.1463

 $\Lambda_{\rm ML} = 30.04 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.4 (a): Formation constants and Gibbs free energies of formation for 1:1 (M/L) complexes in absolute ethanol at 293.15 K.

\(\lambda_{\text{obs}}\) \((\text{cm}^2.\text{Ohm}^{-1})\)	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(/_{M}-/_{obs})$	$\mathbf{K}_{\mathbf{f}}$	$\begin{array}{c} \pmb{\Delta} \; \mathbf{G_f} \\ (\mathbf{k} \; \mathbf{J/mol}) \end{array}$
16.69122	9.91E-05	1.06E-05	14.04614	1319098	34.4584-
16.80059	1.07E-04	2.32E-05	13.93677	600729.6	32.5351-
16.91194	1.15E-04	3.77E-05	13.82542	366337.7	31.3258-
17.02532	1.23E-04	5.43E-05	13.71204	252474.2	30.4156-
17.12624	1.30E-04	7.05E-05	13.61112	193007.8	29.7588-

 $\Lambda_{\rm ML} = 16.58 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.4 (b): Formation constants and Gibbs free energies of formation for 1:1 (M/L) complexes in absolute ethanol at 298.15 K.

∧ _{obs} (cm ² .Ohm ⁻¹)	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	$K_{\rm f}$	$\Delta \ G_f(k \ J/mol)$
23.7688	9.91E-05	1.49E-05	17.2475	1156963.0	-34.7199
23.9220	1.07E-04	3.25E-05	17.0944	526309.1	-32.7611
24.0778	1.15E-04	5.28E-05	16.9385	320589.7	-31.5283
24.2366	1.23E-04	7.60E-05	16.7798	220685.7	-30.5997
24.3779	1.30E-04	9.87E-05	16.6385	168525.9	-29.9291

 $\Lambda_{\rm ML} = 23.618 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.4(c): Formation constants and Gibbs free energies of formation for 1:1 (M/L) complexes in absolute ethanol at 303.15K.

/ _{obs} (cm ² .Ohm ⁻¹)	[L]	(∖obs-∖ML) [L]	$(\land_{M} - \land_{obs})$	K _f	Δ G _f (k J/mol)
26.62185	9.91E-05	2.13E-05	24.99526	1173729.0	-35.3386
26.8406	1.07E-04	4.64E-05	24.77651	533982.5	-33.3471
27.0633	1.15E-04	7.55E-05	24.55381	325306.1	-32.0940
27.29006	1.23E-04	1.09E-04	24.32705	223962.1	-31.1501
27.4919	1.30E-04	1.41E-04	24.12521	171049.6	-30.4686

 $\Lambda_{\rm ML} = 26.407 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.4 (d): Formation constants and Gibbs free energies of formation for 1:1 (M/L) complexes in absolute ethanol at 308.15K.

	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	$\mathbf{K}_{\mathbf{f}}$	$\Delta \ G_f(k \ J/mol)$
31.68424	9.91E-05	2.98E-05	32.64535	1094958.0	-35.7429
31.99048	1.07E-04	6.50E-05	32.33911	497841.6	-33.7170
32.30226	1.15E-04	1.06E-04	32.02733	303088.1	-32.4415
32.61973	1.23E-04	1.52E-04	31.70986	208521.7	-31.4803
32.90230	1.30E-04	1.97E-04	31.42729	159159.1	-30.7859

 $\Lambda_{\rm ML} = 31.383 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.5 (a): Formation constants and Gibbs free energies of formation for 1:2 (M/L) complexes in absolute ethanol at 293.15 K.

/ _{obs} (cm ² .Ohm ⁻¹)	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	K_{f}	$\Delta \; G_f(k\; J/mol)$
17.70214	1.74E-04	1.56E-05	13.03522	836852.7	33.3457-
17.7801	1.80E-04	3.01E-05	12.95726	429811.2	31.7165-
17.8725	1.87E-04	4.86E-05	12.86486	264722.3	30.5314-
17.9665	1.94E-04	6.87E-05	12.77086	186021.2	29.6687-
18.04839	2.00E-04	8.72E-05	12.68897	145592.5	29.0695-

 $\Lambda_{\rm ML} = 17.61 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.5 (b): Formation constants and Gibbs free energies of formation for 1:2 (M/L) complexes in absolute ethanol at 298.15K.

	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	$\mathbf{K_f}$	$\Delta \ G_f(k \ J/mol)$
25.17068	1.74E-04	2.34E-05	15.84576	678141.2	-33.3915
25.28761	1.80E-04	4.52E-05	15.72883	347832.1	-31.7311
25.42621	1.87E-04	7.29E-05	15.59023	213868.5	-30.5216
25.56722	1.94E-04	1.03E-04	15.44922	150020.1	-29.6398
25.69005	2.00E-04	1.31E-04	15.32639	117235.2	-29.0266

 $\Lambda_{\rm ML} = 25.036 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.5(c): Formation constants and Gibbs free energies of formation for 1:2 (M/L) complexes in absolute ethanol at 303.15K.

	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	$\mathbf{K_f}$	$\Delta \ G_f(k \ J/mol)$
28.7905	1.74E-04	3.05E-05	22.82661	747932.8	-34.1991
28.9659	1.80E-04	6.31E-05	22.65121	358723.1	-32.3412
29.1738	1.87E-04	1.04E-04	22.44331	214816.0	-31.0447
29.38532	1.94E-04	1.49E-04	22.23179	148784.6	-30.1160
29.56956	2.00E-04	1.91E-04	22.04755	115497.5	-29.4756

 $\wedge_{\rm ML} = 28.615 \text{ cm}^2.\text{Ohm}^{-1}.$

Table.5 (d): Formation constants and Gibbs free energies of formation for 1:2 (M/L) complexes in absolute ethanol at 308.15K.

	[L]	$(\land_{obs} - \land_{ML})$ [L]	$(\land_{M} - \land_{obs})$	K _f	$\Delta \ G_f(k \ J/mol)$
34.7881	1.74E-04	5.45E-05	29.5414	541817.3	-33.9346
35.0609	1.80E-04	1.06E-04	29.2686	277390.2	-32.2138
35.3843	1.87E-04	1.70E-04	28.9452	170172.5	-30.9579
35.7134	1.94E-04	2.40E-04	28.6161	119090.1	-30.0405
36.0000	2.00E-04	3.05E-04	28.3295	92871.11	-29.4013

 $\Lambda_{\rm ML} = 34.475 \text{ cm}^2.\text{Ohm}^{-1}$

Table.6: The enthalpies (ΔH_A) and entropies (ΔS_A) of CuCl₂ with L2 at different temperatures.

M:L	$(\Delta S_A)kJ/mol.K$	$(\Delta S_A)kJ/mol.K$			
Temp	293.15 K	298.15 K	303.15 K	308.15 K	kJ/mol
2:1	0.3594	0.3559	0.3584	0.3586	55.9789
1:1	0.3568	0.3534	0.3558	0.3561	54.1697
1:2	0.3403	0.3374	0.3395	0.3397	47.9787

Table.7: The enthalpies (ΔH_f) and entropies (ΔS_f) of CuCl₂ with L2 at different temperatures.

M:L	(ΔS_f) kJ/mol.H	$(\Delta S_f)kJ/mol.K$			
Temp	293.15 K	298.15 K	303.15 K	308.15 K	
2:1	0.3049	0.3008	0.2979	0.2951	53.5394
1:1	0.2877	0.2837	0.2811	0.2779	49.8936
1:2	0.2350	0.2743	0.2724	0.2672	48.4034

The enthalpy change(ΔH) and the entropy change($T\Delta S$) evaluated for CuCl₂-ligand complexes indicate that (L1) may act as flexidentate ligand.

The Enthalpy change of association (ΔH_A), and the Enthalpy change of formation (ΔH_f) of CuCl₂ in absolute ethanol at 293.15, 298.15, 303.15 and 308.15 K estimated by plotting log K_A and log K_f for different M/L concentrations for (2:1), (1:1) and (1:2) stoichiometric complexes against 1/T are represented in (figs. 7,8).

II. CONCLUSION

This work concentrated on the behavior of CuCl2 with the ligand conductometrically .The main target is to discuss the complexation between the metal and ligand for evaluating different concentrations from the metal ion in different solutions.

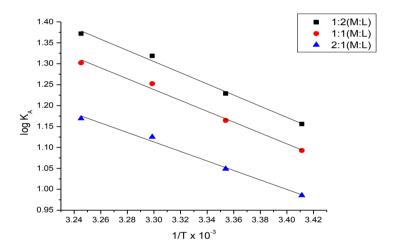


Fig. 7 The relation between log (K_A) for (2:1), (1:1) and (1:2) stoichiometric complexes against 1/T.

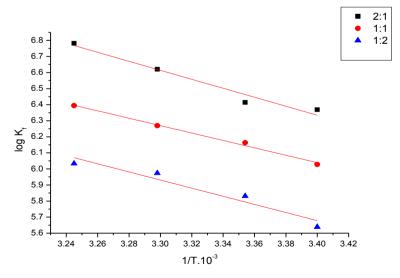


Fig. 8 The relation between $log(K_f)$ for (2:1), (1:1) and (1:2) stoichiometric complexes against 1/T.

The Enthalpy change (ΔH) of $CoCl_2$ in absolute ethanol was calculated using the equation,

Slope =
$$\frac{-\Delta H}{2.303 \text{ x R}}$$

Where R is the real gas constant.

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