International Journal of Chemical Science
Online ISSN: 2523-2843, Print ISSN: 2523-6075; Impact Factor: RJIF 5.22
Received: 10-05-2020; Accepted: 25-05-2020; Published: 12-06-2020
www.chemicaljournals.com
Volume 4; Issue 3; 2020; Page No. 06-11

Cyclovoltammetric data for the interaction of thallous chloride with di ethyl amino ethyl cellulose (DEAEC) in HNO_3 using glassy carbon electrode

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Abstract
The redox behavior data for thallous chloride using 0.1M HNO_3 in absence and presence of polymer di ethyl amino ethyl cellulose were evaluated cyclic voltammetrically. Cyclic voltammetry of thallous chloride (TlCl) was achieved using glassy carbon electrode and in presence of di ethyl amino ethyl cellulose (DEAEC) as natural polymer. The cyclic voltammetry and the kinetic data for the redox reactions of TlCl was studied and their values were discussed. The thermodynamic stability constants and Gibbs free energies of interaction between TlCl and DEAEC were evaluated and their values were discussed.

Keywords: cyclic voltammetry, thallous chloride, di ethyl amino ethyl cellulose, kinetic parameters, thermodynamic parameters

1. Introduction

Thallous chloride known as thallium (I) chloride, it is created from the reaction of thallium (I) sulfate with hydrochloric acid. It is used as a diagnostic radiopharmaceutical. It used for diagnosis of heart and parathyroid problems. Other side effects as: blurred vision, chest pain or discomfort, chills, confusion, cough difficulty with breathing, difficulty with swallowing and dizziness [1]. Thallous chloride without carrier accumlates in the heart (myocardium) in a similar way to that of potassium. Thallous chloride have been found to visualize areas of interaction or no labeled regions which are confirmed by electrocadiographic and enzyme changes. Also regions of transient myocardial ischemia corresponding to areas. Prefer used by coronary arteries with partial stenoses can be visualized by thallous chloride [2, 3]. Therefor estimation of different redox behavior of TlCl is important for following its amounts.

2. Experimental

Thallous chlorid of the type…. was used. Di ethyl amino ethyl cellulose and hydrochloric acid of the type was provided from Al Gomhoria pharmaceutical company. The electrochemical study was carried out by usind DY2000 potentiostat in cell containing 30 ml of 0.1 HCl supporting electrode. The three used electrodes are, pt (platinum wire) count's electrode, silver- silver chloride in saturated KCl used as reference electrode and glassy carbon electrode as working electrode. The cell used is: $Ag/AgCl_{(s)}$, $KCl_{(sat)}$ | aqueous 0.1M HCl | GCE. It is very important to clean the working electrode with fine emery paper doped with Al_2O_3 in manufacture made. Then the working electrode was washed several times with distilled water and dried.

3. Results and Discussion

The electrochemical behavior of thallous chloride (TlCl) alone in 0.1 M HCl at 289.15K. Electrochemical behavior of thallous chloride (TlCl) alone in 0.1 M HCl using window range from 1.5V to ~ 1.5V. Two main peaks were obtained for TlCl, one for the reduction process at ~ -0.1V and the other in the oxidation site at ~0.9V. Quasirversible study was suggested for the redox reaction of TlCl, see Fig. (1).

The reduction and oxidation process of Tl alone took place by consuming one electron. The reduction mechanism for TlCl was found to go throw the following equation:

$$Tl^+ + e^- \rightleftharpoons Tl_{(s)} \qquad \text{at} \sim -0.1V \text{ ......} \qquad (1)$$

Whereas the oxidation process took place following equations (2) and/or equation (3).

$$Tl_{(s)} \rightleftharpoons Tl^+ + e^- \qquad \text{at} \sim -0.9V \text{ ......} \qquad (2)$$

$$Tl_{(s)} + Cl^- \rightleftharpoons TlCl_{(s)} + e^- \qquad \text{at} \sim -0.9V \text{ ......} \qquad (3)$$

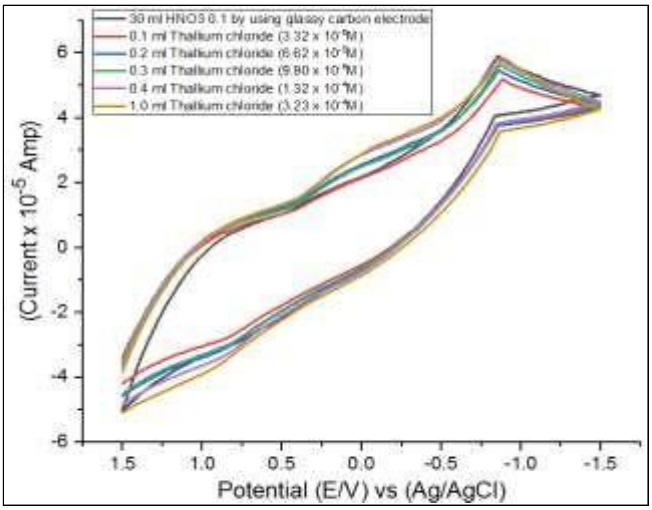

Fig 1: Effect of different concentrations of Thallium chloride in 0.1M HNO3 using glassy carbon electrode at 289.15 Kand scan rate $0.1V.S^{-1}$.

We noticed that both redox waves for TlCl are broad peaks. That is to say, wide range peaks. Proceeding the reduction of TlCl by two mechanisms, i.e. two parallel ways, following equations (1) and (4) from a wide range (0.2- -0.3V) of reduction as:

$$TlCl_{(s)} + e^- \rightleftharpoons Tl_{(s)} + Cl^- \quad \text{at} \sim -0.1V \ \ldots\ldots \qquad (4)$$

Also the oxidation of TlCl happened by two parallel process following equations 2 and 3 resulting in abroad oxidation peak in range (0.7 – 1.0V) Analysis of the cyclovoltammetric TlCl waves: The different equations used for the electrochemical results by cyclic voltammetry are [15-25].

$$I_P = [0.4463\, n^{3/2} F^{3/2} D^{1/2} A\, C / (RT)^{1/2}]\, v^{1/2} \ \ldots\ldots \qquad (5)$$

$$D^{1/2} = (\text{slope}, I_P \text{ vs } v^{1/2}) \text{ x } ((RT)^{1/2} / 0.4463\, n^{3/2} F^{3/2} D^{1/2} A\, C) \ \ldots\ldots \qquad (6)$$

$$\Delta E_P = E_{Pa} - E_{Pc} = 2.303(RT/n\,F) \ \ldots\ldots \qquad (7)$$

$$\Psi = \gamma^{\alpha} K_s / \sqrt{\pi (n\,F/RT)\, v\, D_a} \ \ldots\ldots \qquad (8)$$

$$\gamma = \sqrt{D_a / D_c} \ \ldots\ldots \qquad (9)$$

Where Ψ is the charge transfer parameter taken as one for better approximation [13], α is the charge transfer coefficient, Ks is the standard rate constant for electron transfer, ν is the scan rate, D_a is the anodic diffusion coefficient, D_c the cathodic diffusion coefficient, F faraday constant, T is the absolute temperature, n is the number of electrons and α = 0.5 for reversible calculations A is the area of the working electrode. The ks which is the heterogenous electron rate constant can be evaluated by applying equation (10) [31, 36].

$$K_s = 2.18\, (D\, \alpha n_a F\, v / RT)^{1/2} \exp \{\alpha 2\, n\, F\, (E_{pc} - E_{PA})/RT\} \ \ldots\ldots \qquad (10)$$

The electron surface coverage was estimated for both reduction and oxidation process by applying equation (11) [30-35].

$$\Gamma = I_P\, 4RT/n^2 F^2 A\, v \ \ldots\ldots \qquad (11)$$

The quantity of the electricity used for the redox reactions was estimated on applying equation (12) [33]

$$Q = n\, F A\, \Gamma \ \ldots\ldots \qquad (12)$$

All the above parameters following equations (5 - 11) for redox behavior of TlCl alone in 0.1 M HCl were calculated and in Table (1) cited.

Table 1: Effect of different concentrations of Thallium chloride by using GCE at 289.15K and scan rate $0.1 V.S^{-1}$.

[M] x10^5 mol.L^{-1}	**Ep,a Volt**	**Ep,c Volt**	**ΔEp Volt**	**(-) Ip,a x10^6 Amp**	**Ip,c x10^6 Amp**	**Ip,a/Ip,c**	**E° Volt**	**Da x10^{10} $cm^2.s^{-1}$**	**Dc x10^{10} $cm^2.s^{-1}$**	**αnac**	**Ksc x10^5**	**Γ c x10^9 mol.cm^{-2}**	**(+) Qc x10^6 C**	**Γ a x10^9 mol.cm^{-2}**	**(-) Qa x10^6 C**
3.32	0.803	0.014	0.789	1.39	3.70	0.374	0.408	2.375	16.9	0.522	3.59	1.218	3.69	0.456	1.38
6.62	0.823	0.029	0.794	2.40	5.06	0.474	0.426	1.789	7.97	0.439	2.35	1.667	5.05	0.789	2.39
9.90	0.828	0.035	0.793	3.50	5.12	0.683	0.431	1.699	3.64	0.441	1.58	1.683	5.10	1.149	3.48
13.2	0.831	0.031	0.799	4.15	6.13	0.678	0.431	1.357	2.96	0.436	1.52	2.016	6.11	1.366	4.14
32.3	0.928	-0.033	0.961	5.37	6.58	0.815	0.447	0.377	0.568	0.795	4.53	2.166	6.56	1.766	5.35

It was observed from Table (1) the following findings

1. The redox reaction of TlCl is quasireversible one because ΔE_p is greater than 0.59 V
2. The cathodic peak are mora developed than the anodic peak with greater limiting current than the oxidation one
3. The cathodic diffusion coefficient D_c is greater than is the anodic diffusion coefficient D_a indicating the case of formation of the reduction peak than that of the oxidation
4. The electrode surface coverage for the reduction peak Γ_c is greater than that for the oxidation peak Γ_a indicating more facility reduction to proceed oxidation for TlCl solutions in 0.1M HCL.
5. The electrode quantity of electricity Q_C for the cathodic peak is greater than Q_a for the anodic peak indicating that the redox reaction is mora reduction favourable.
6. The ks, heterogenous electron rate constant is increase firstly by increase of TlCl concentration from 3.32 x 10^{-5} to 6.62 x 10^{-5} due to the high increase in both I_{Pc} and I_{Pc}. Also the increase in Ks by the use of TlCl concentration 3.32x10^{-5}M is because the rush increase in both I_{Pa} and I_{Pc}. Decrease of Ks on using TlCl concentration of 9.9 x 10^{-3} and 3.32 x 10^{-3} due to small increase in both I_{Pa} and I_{Pc}.
7. All parameters given in Table (1) are approximately increase by the increase of TlCl concentration confirm the diffusion mechanism.

Effect of different scan rates, 0.1, 0.05, 0.02, 0.01 V/S were studied for electrochemical behavior of 3.23 x 10^{-5} M TlCl.

Fig (2) show the different cyclic voltammograms for TlCl at different scan rate giving no new peaks. Fig (2) the relation between peak current and $\nu^{1/2}$, two straight lines for the oxidation and reduction peaks with slope ~ 0.5 indicating the diffusion controlled reaction.

All the data given in Table (2) are increased by the decrease of scan rate other than Ks values. This indicate the diffusiability of the reactions.

The electrochemical behavior for TlCl in presence of 1% (W/W) di ethyl amino ethyl cellulose.

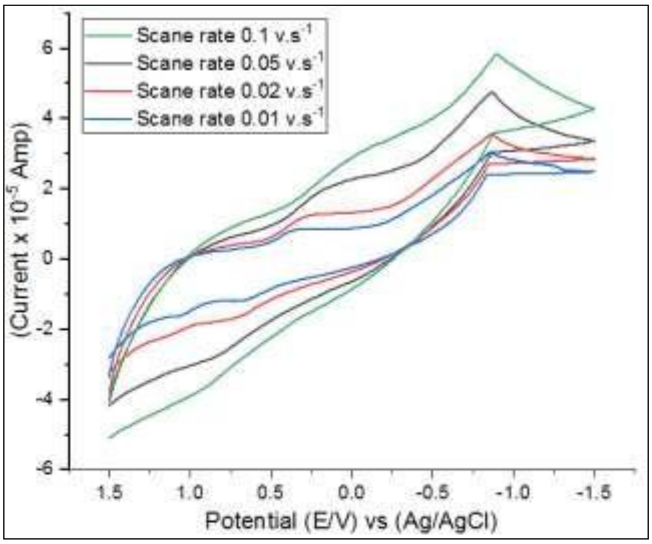

Fig 2: Effect of different scan rates of (3.23x10^{-4} M) Thallium chloride by using glassy carbon electrode at 289.15 K.

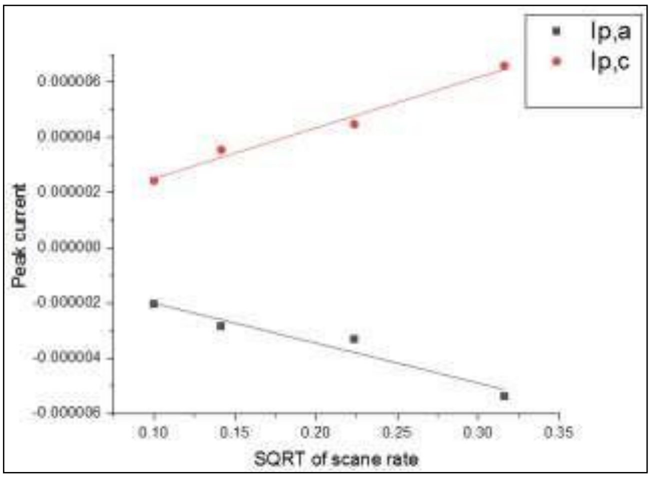

Fig 3: The relation between peak current and SQRT of scan rate for of Thallium chloride alone.

Fig. (2) Represent the electrochemical behaviorof the comlex formed by the interaction of TlCl with (DEAEC) in 0.1 M HCl at 289.15 K.

Table 2: Effect of different scan rates of (3.23x10^{-4} M) Thallium chloride by using GCE at 289.15K

ν V.S^{-1}	Ep,a Volt	Ep,c Volt	ΔEp Volt	(-)Ip,a x10^{6} Amp	Ip,c x10^{5} Amp	Ip,a/Ip,c	E° Volt	Da x10^{13}cm^{2}.s^{-1}	Dc x10^{12} cm^{2}.s^{-1}	αna	ksc x10^{3}	Γ c x10^{9} mol.cm^{-2}	(+)Qc x10^{6}C	Γ ax10^{9} mol.cm^{-2}	(-) Qa x10^{6}C
0.1	0.928	-0.033	0.961	5.37	6.58	0.815	0.447	3.77	5.678	0.795	45.3	2.166	6.56	1.766	5.35
0.05	0.821	0.1964	0.624	3.30	4.47	0.738	0.509	2.85	5.235	0.808	1.06	2.941	8.91	2.172	6.58
0.02	0.692	0.337	0.355	2.83	3.54	0.799	0.514	5.25	8.208	0.736	0.0536	5.823	17.6	4.659	14.1
0.01	0.627	0.373	0.253	2.03	2.43	0.837	0.500	5.40	7.714	0.819	0.0139	7.984	24.2	6.679	20.2

Same window range from 1.5 ~ -1.5V was used the cyclic voltammograms seen in Fig (2) and data given in Table (2) show the following

1. I$_{Pa}$ and I$_{Pc}$ for the different concentration of di ethyl amino ethyl cellulose are less than that on using 3.23 x 10^{-4} M TlCl alone indicating complex reaction.
2. D$_a$ and D$_c$ values are greater than that of 3.32 x 10^{-4} M TlCl alone indicating more complexation by more diffusion.
3. Γ$_a$ and Γ$_c$ are small in presence of di ethyl amino ethyl cellulose than in absence of it indicating also complexation behavior
4. All the heterogenous rate constant values ks are small than 3.23 x10^{-4} M TlCl alone favouring attraction by complexation.

Table 3: Effect of different Di ethyl amino cellulose 1% by using GCE at 289.15K

[M] x10^{4} mol.L^{-1}	[L] x10^{4} mol.L^{-}	Ep,a Volt	Ep,c Volt	ΔEp Volt	(-) Ip,a x10^{6} Amp	Ip,c x10^{6} Amp	Ip,a/Ip,c	E° Volt	Da x10^{12} cm^{2}.s^{-1}	Dc x10^{11} cm^{2}.s^{-1}	αn$_a$c	ksc x10	Γ c x10^{9} mol.cm^{-2}	(+) Qc x10^{5} C	Γ a x10^{10} mol.cm^{-2}	(-) Qa x10^{6} C
3.23	0.800	0.941	0.089	0.852	3.36	6.41	0.525	0.515	14.816	5.38	0.067	1.37	2.109	6.39E-06	11.06	3.35
3.23	0.958	0.935	0.138	0.797	3.21	5.23	0.614	0.537	13.518	3.59	0.0263	1.03	1.721	5.21	10.57	3.20
3.23	1.12	0.903	0.081	0.822	2.67	4.28	0.623	0.492	9.335	2.40	0.072	0.656	1.409	4.27	8.782	2.66
3.23	1.27	0.902	0.099	0.802	2.20	3.14	0.700	0.501	6.321	1.29	0.034	0.575	1.032	3.13	7.227	2.19
3.23	1.43	0.946	0.075	0.871	1.65	2.88	0.574	0.511	3.585	1.09	0.024	1.26	0.947	2.87	5.442	1.65
3.23	1.59	0.921	0.077	0.844	1.19	2.53	0.471	0.499	1.854	0.838	0.031	0.736	0.832	2.52	3.914	1.19

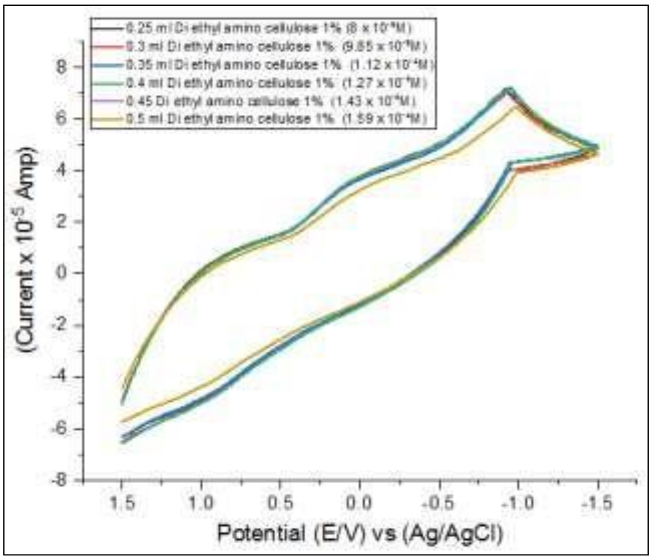

Fig 4: Effect of different concentrations of Di ethyl amino cellulose 1% on potassium permanganate by using glassy carbon electrode at 289.15 K and scan rate 0.1 V.S^{-1}.

Effect of different scan rates on TlCl + di ethyl amino ethyl cellulose

Effect of different scan rates was studied for interaction of TlCl with di ethyl amino ethyl cellulose and cyclic voltammetry data was observed in Table (5) with smaller value than that for TlCl alone indicating the complexation reaction.

The complexation stability constant (β) for TlCl – DEAEC complexes in 0.1 M HCl is the application of equation (13) [27, 36]

$$(E_P)_C - (E_P)_M = 2.303(RT/nF) * (\log \beta_C + 2.303 (RT/nF) \log C_L) \ldots\ldots \quad (13)$$

Where β_c is the stability constant, $(E_P)_M$ is the peak potential of TlCl in absence of DEAEC, $(E_P)_C$ is the peak potential for the complex. Other symbols are explained before.

The Gibbs free energies of interaction between TlCl and DEAEC were calculated [19, 26] by using equation (14) [30, 39].

$$\Delta G = -2.303\ RT \log \beta_{MX} \quad \ldots\ldots\ldots\ldots \quad (14)$$

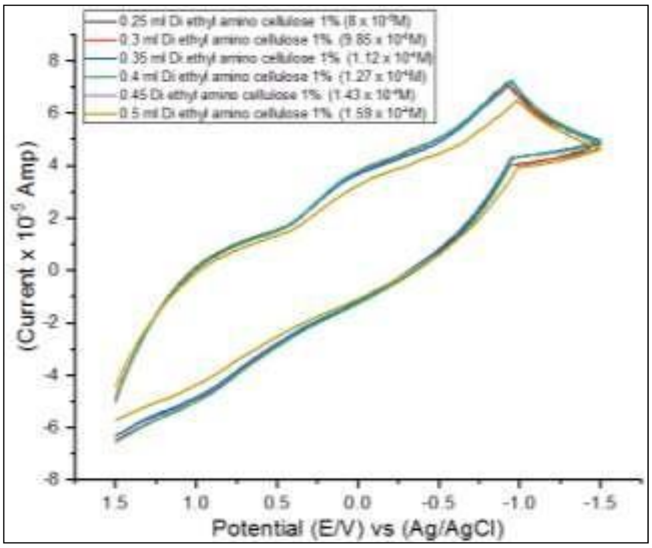

Fig 4: Effect of different concentrations of Di ethyl amino cellulose 1% on potassium permanganate by using glassy carbon electrode at 289.15 K and scan rate 0.1 V.S^{-1}.

Table 4: Effect of different scan rates of in the presence of Di ethyl amino cellulose 1% by using GCE at 289.15 K.

ν V.S^{-1}	Ep,a Volt	Ep,c Volt	ΔEp Volt	(-)Ip,a x10^7 Amp	Ip,c x10^5 Amp	Ip,a/Ip,c	E° Volt	Da x10^{14}cm^2.s^{-1}	Dc x10^{12} cm^2.s^{-1}	αna	ksc x10^4	Γ c x10^9 mol.cm^{-2}	(+)Qc x10^6 C	Γ ax10^9 mol.cm^{-2}	(-) Qa x10^6C
0.1	0.921	0.077	0.844	11.9	2.53	0.471	0.499	2.101	0.949	1.493	78.3	0.832	2.52	0.391	1.19
0.05	0.730	0.217	0.514	9.07	2.49	0.364	0.474	2.440	1.838	0.898	2.18	1.637	4.96	0.597	1.81
0.02	0.659	0.346	0.313	6.71	2.05	0.327	0.503	3.341	3.131	1.160	0.273	3.379	10.2	1.104	3.34
0.01	0.598	0.413	0.184	5.74	1.37	0.420	0.506	4.897	2.774	1.999	0.065	4.498	13.6	1.889	5.73

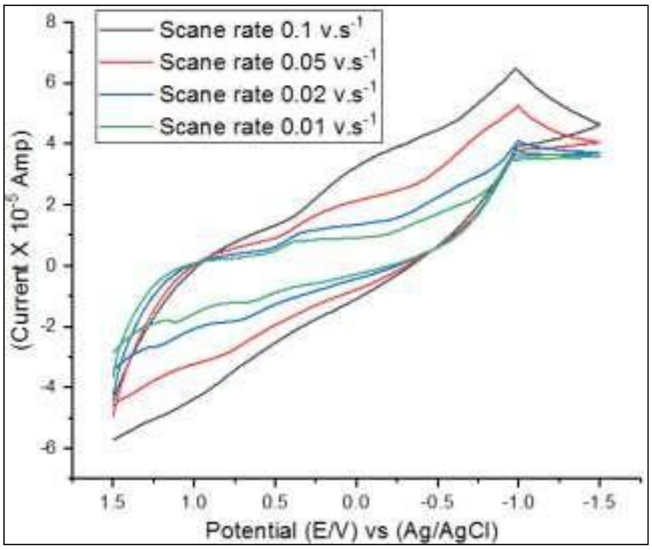

Fig 5: Effect of different scan rates of in the presence of Di ethyl amino cellulose 1% by using glassy carbon electrode at 289.15 K.

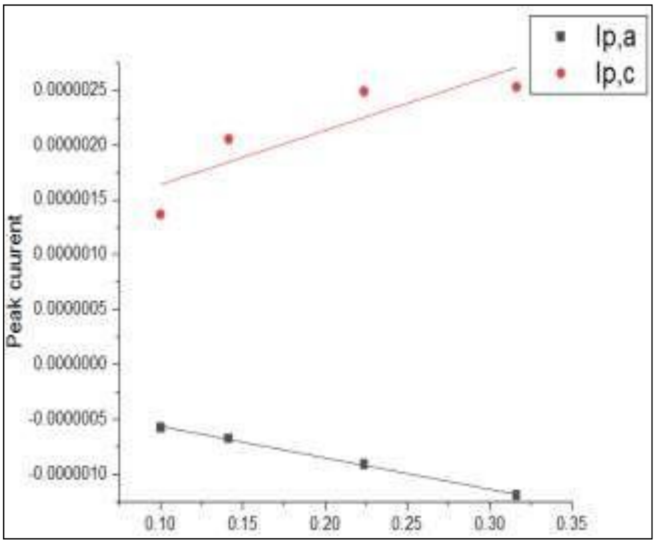

Fig 6: The relation between peak current and SQRT of scan rate in presence of Di ethyl amino cellulose 1%

Table 5: Stability constant for (Thallium chloride - Di ethyl amino cellulose) complex.

[M] x10^4 mol.L^{-1}	[L] x 10^4 mol.L^{-1}	(Ep,1/2)M	(Ep,1/2)C	ΔE v	J (L/J)	Log βj	ΔG (KJ/mol)
3.20	0.800	0.447	0.515	-0.068	0.25	0.372	0.899
3.19	0.958	0.447	0.537	-0.089	0.3	0.284	1.955
3.19	1.12	0.447	0.492	-0.045	0.35	0.301	-3.289
3.18	1.27	0.447	0.501	-0.054	0.4	0.258	-3.459
3.18	1.43	0.447	0.511	-0.063	0.45	0.0735	-3.458

From the data given in Tables (3-5), were conclude that the interaction between TlCl and DEAEC leads to formation of strong complex. Also the Gibbs free energies of interaction between TlCl and DEAEC support the complexation reaction between the two

Conclusion

We explained the redox behavior for thallous chloride from cyclic voltammetric data in 0.1 M HCl using glassy carbon electrode at 289.15 K.

Effect of di ethyl amino ethyl cellulose was studied and their effect were interpreted.

The complex formation between thallous chloride and di ethyl amino ethyl cellulose was found to happen and Gibbs free energies were obtained explaining strong complexation interaction.

Declaration of Interests

The authors declare that they have no conflict with anybody about this work.

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