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Solvation Cyclic voltammetry Parameters for Na_2WO_4 in $NaClO_4$, HCl Media and Interaction with Congo Red Dye

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Abstract

Nanotechnology has been a promising technology over the years. Carbon nanotubes (CNTs) are one of the principal technologies that are The study examined the effects of different concentrations of sodium tungstate (Na_2WO_4) using cyclic voltammetry with a glassy carbon electrode (EGC). In acidic HCl and neutral $NaClO_4$ media, the cathodic reaction of sodium tungstate was characterized by the transfer of tungstate ions to WO_2. Additionally, the oxidation of WO_2 ions to WO_3 was observed. Cyclic voltammetry parameters for sodium tungstate in the absence and presence of Congo red dye were analyzed in both $NaClO_4$ and HCl media. The stability constants and Gibbs free energies of complexation were estimated for both media, revealing higher values in $NaClO_4$ compared to HCl. This suggests that the complexation reaction between Na_2WO_4 and Congo red dye is more favorable in $NaClO_4$ than in HCl. Furthermore, the kinetic and solvation parameters for the interaction of Na_2WO_4 with Congo red in 0.1 M $NaClO_4$ were lower than those observed in 0.1 M HCl, attributed to a faster electron transfer rate in the HCl medium.

Highlights

1. Study focused on sodium tungstate (Na_2WO_4) using cyclic voltammetry with a glassy carbon electrode.
2. Cathodic reaction involved transfer of tungstate ions to WO_2 in acidic HCl and neutral $NaClO_4$.
3. Observed oxidation of WO_2 ions to WO_3.
4. Cyclic voltammetry parameters analyzed with and without Congo red dye in both media.
5. Stability constants and Gibbs free energies of complexation were greater in $NaClO_4$ than in HCl.
6. Complexation between Na_2WO_4 and Congo red dye is more favorable in $NaClO_4$.
7. Kinetic and solvation parameters indicated faster electron transfer in HCl compared to $NaClO_4$.

1. Introduction

The electrochemical behavior of the sodium tungstate ion WO_4^{2-} in acidic media attracted attention due to its use in technological applications as tungstate films [1–3]. The application of WO_4-films as semiconducting materials for photochemical analysis and hydrogen production from water [4,5]. Also, tungstate is used as an electrochemical material for polishing [6–8], and its electrochemistry is used to explain the dielectric properties of tungstate WO3 films [8–20]. The electro redox reactions of sodium tungstate have several benefits. The electrochemical behavior of the sodium tungstate ion WO $^{-2}$ in acidic media attracted attention due to its use in technological applications as tungstate films [1–3]. The application of WO^{4-} films as semiconducting materials for photochemical analysis and hydrogen production from water [4,5]. Also, tungstate is used as an electrochemical material for polishing [6–8], and other applications.Different concentrations of sodium tungstate (Na_2WO_4) were investigated using cyclic voltammetry with a glassy carbon electrode (EGC). Previous studies have examined the electrochemical behavior of tungstate ions, suggesting that the cathodic reaction in acidic HCl and neutral $NaClO_4$ media involves the transfer of tungstate ions to WO_2. The oxidation of WO_2 ions to WO_3 was also observed in these conditions.The goal of this work is to explore the solvatochromic cyclic voltammetry parameters for sodium tungstate in the absence and presence of Congo red dye in both $NaClO_4$ and HCl media. Stability constants and Gibbs free energies of complexation were estimated for both media, revealing that complexation is more pronounced in $NaClO_4$ compared to HCl. This supports the idea that the interaction between Na_2WO_4 and Congo red dye is stronger in $NaClO_4$ solutions. Additionally, the kinetic and solvation cyclic parameters for the interaction of Na_2WO_4 with Congo red in 0.1 M $NaClO_4$ were found to be lower than those in 0.1 M HCl, attributed to a faster electron transfer rate in the HCl medium.

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electrochemistry is used to explain the dielectric properties of tungstate WO_3 films [18–20]. The electro redox reactions of sodium tungstate have several benefits. Firstly, it is used for tungsten metal production with high purity from sodium tungstate, since tungsten is a critical strategic metal with applications in electronics and aerospace. Secondly, energy efficiency, since the redox processes are generally more energy-efficient compared to traditional pyro metallurgical methods for extracting tungsten from ores. This can lead to lower energy costs. Thirdly, selectivity, since electro reduction allows for the selective reduction of tungsten ions, separating them from other metalions that may be present in the feedstock. This can result in a purer final tungsten product. Fourthly, waste reduction, since electrolysis can minimize waste generation compared to some other tungsten extraction techniques. The electrolyte can often be reused, reducing the amount of waste produced. Fifthly, process flexibility, since electro reduction can be carried out at relatively low temperatures, making it more flexible than high- temperature pyrometallurgical processes. Sixthly, environmental benefits, since by being more energy- efficient and generating less waste, the electro reduction of sodium tungstate can have a lower environmental impact compared to other tungsten production methods. Therefore, giving a lot of data about sodium tungstate in different media and interaction with some reagents are necessary for understanding the different tungstate behaviors.

2. Materials &Method

The materials (sodium tungstate, sodium perchlorate, and HCl) were supplied by Sigma-Aldrich. Congo Red, High Purity, 3,3′-[(1,1′-Biphenyl)-4,4′-diylbis(azo)]bis(4-amino-1-naphthalenesulfonic Acid) 2Na from Dr.Theodor Schuchardt.in Munchen. The water was deionized. Using a three-electrode setup, cyclic voltammetry measurements were carried out with the DY2000 (USA). The reference electrode (Ag/AgCl/saturated KCl), the working electrode (glassy carbon electrode), and the auxiliary electrode (Pt electrode) are the various electrodes.

3. Results &discussion

3.1. *Cyclic voltammogram for different concentrations of Na2WO4 in NaClO4 (0.1) M*

The cyclic voltammetry was performed for 0.1M $NaClO_4$ solutions and by adding different concentrations of sodium tungstate (0.1M) using the glassy carbon electrode as the electrode in use (EGC). The waves when the EGC is present are in the window range of 1.5V to -1.5V, and one main reduction peak at -0.1V and one main oxidation peak at +0.4V are formed. As shown in Figure (1) and Table (1), an increase in the cathodic and oxidation waves was seen by adding numerous Na_2WO_4 (0.1M) concentrations, suggesting a diffusion-controlled process. All the parameters shown in Table 1 are increased by increase of tungstate concentration and this confirm the diffusibility of the redox reaction under consideration [20-22].

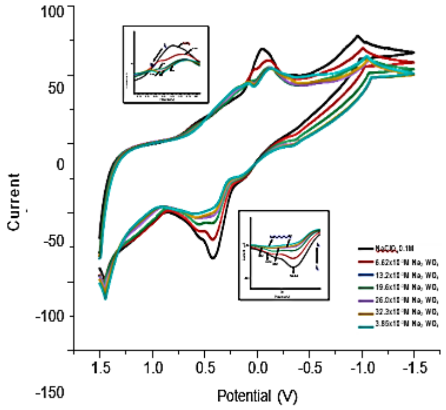

Fig. 1. A Cyclic voltammograms for different concentrations of Na2WO4 in NaClO4 (0.1) M at scan rate 0.1V/s and 292.65K.

The cathodic reaction of sodium tungstate in acidic and neutral media was suggested as transfer of tungstate ion to WO_3 as [13]:

$$WO_4^{2-} + H^+ \leftrightarrow WO_3 + OH^-$$

The formed WO_3 was reduced to WO_2 and W_2O_5 consuming two electrons as explained in the following equations:

$WO_3 + 2H^+ + 2e^- \leftrightarrow WO_2 + 2H_2O$ at ~ -0.1 V

$2WO_2 + 2H^+ + 2e^- \leftrightarrow W_2O_5 + H_2O$

The last W_2O_5 was also reduced to WO_2 as:

$W_2O_5 + 2H^+ + 2e^- \leftrightarrow 2WO_2 + H_2O$ at ~ -0.2 V

The oxidation of WO_2 to WO_3 was observed at +0.3V.Whereas the oxidation of WO_2 to W_2O_5 was found to be at +0.5V.We select the primary oxidation peak at 0.1V and the primary reduction peak at -0.1V.

Table 1 A :Effect of different concentrations of Na2WO4 by using EGC at scan rate 0.1V/s and 292.65K.

[M] x10^3 mol.L^{-1}	**Volt**			**Amp**		**Ip,a/Ip,c**	**Volt**
	(-) Ep,a(V)	Ep,c(V)	ΔEp(V)	(-)Ip,ax10^6	Ip,cx10^6		E°
6.62	0.395	0.108	0.503	37.97	32.81	1.157	0.144
13.2	0.416	0.108	0.524	26.88	17.84	1.506	0.154
19.6	0.414	0.078	0.491	34.14	22.50	1.517	0.168
26.0	0.449	0.071	0.519	25.01	25.44	0.983	0.189
32.3	0.549	0.076	0.624	25.47	27.89	0.913	0.236

Table 1 B : Continue.

Dax10^5 cm^2.s^{-1}	**Dc x10^5 cm^2.s^{-1}**	**Epc/2**	**Epa-Epc/2**	**αna**	**ksc x10^2 cm^2.s^{-1}**	**Γc x10^9 mol.cm^{-2}**	**(+)Qcx10^5 Columb**	**Γax10^9 mol.cm^{-2}**	**(-)Qax10^5 Columb**
5.667	4.23	0.060	0.1679	0.279	3.22	2.730	1.65	3.160	1.91
7.194	3.17	0.069	0.1776	0.264	1.31	1.485	9.00	2.237	1.36
5.226	2.27	0.071	0.1483	0.316	6.31	1.872	1.13	2.841	1.72
1.598	1.65	0.209	0.2793	0.168	6.86	2.117	1.28	2.081	1.26
1.075	1.288	0.226	0.3017	0.155	4.65	2.321	1.41	2.12	1.28
4.698	6.871	0.127	0.2428	0.193	4.25	2.021	1.22	1.67	1.01

3.2. *Evaluation of different solvation parameters for sodium tungstate in the absence and presence of congo red in NaClO4 media:*

Table 1A ,1B show various solvation, kinetic, and thermodynamic parameters were evaluated as explained in previous works, and the results are presented also in other Tables in Tables 2 A, 2 B , 3, with high Gibbs free energies indicating a good complexion reaction. The cyclic voltammograms are given in Fig. 1B for the effect of congo red dye on the redox reaction of sodium tungstate. Showing the same wave but with a decrease in their currents, informing a complexation reaction between the two in Figure (2) and Tables 2 A, 2B.

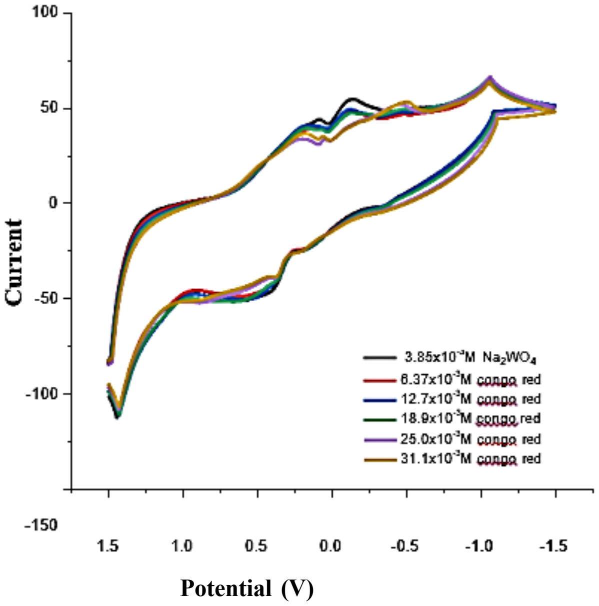

Fig. 2. Cyclic voltammogram for the interaction of (32.2x10- 3M) Na2WO4 and different concentrations of congo red dye at scan rate 0.1V/s and 292.65K

Table 2 A :Effect of different concentrations of Congo red by using EGC at scan rate (0.1 V/s and 292.65K

[L]x10^3 mol.L^{-1}	**Volt**			**Amp**		**Ip,a/Ip,c**	**volt**
	(-) Ep,a(V)	Ep,c(V)	ΔEp(V)	(-)Ip,ax10^6	Ip,cx10^6		E°
6.37	0.504	0.114	0.617	13.43	27.14	0.494	0.195
12.7	0.495	0.078	0.572	14.95	22.07	0.677	0.209
18.9	0.541	0.079	0.619	21.28	20.35	1.045	0.231
25.0	0.363	0.426	0.788	20.26	89.49	2.264	0.031
31.1	0.359	0.464	0.822	14.84	11.14	1.331	0.053

Table 2 B :Continue

Dcx10^5 cm^{-2}.s^{-1}	**Epc/2**	**Epa-Epc/2**	**αna**	**ksc x10^2 cm^{-2}.s^{-1}**	**Γc x10^9 mol.cm^{-2}**	**(+)Qcx10^5 columb**	**Γa x10^9 mol.cm^{-2}**	**(-)Qax10^5 columb**
4.23	0.060	0.16797	0.279	3.22	2.7309	1.65	3.160	1.91
3.17	0.069	0.1776	0.264	1.31	1.4851	9.00	2.2371	1.36
2.27	0.071	0.1483	0.316	6.31	1.8726	1.13	2.841	1.72
1.65	0.209	0.2793	0.168	6.86	2.1174	1.28	2.081	1.26
1.288	0.226	0.3017	0.155	4.65	2.3210	1.41	2.12	1.28

Table 3 Stability constants and Gibbs free energies of solvation for (Na2WO4+Congo red) interaction in NaClO4 medium

[L]x10^3 mol.L^{-1}	**Metal** (Ep,a)M	**Complex** (Ep,a)C	**ΔE mv**	**log βj**	**ΔG (kJ /mol)**
6.58	0.200	0.195	0.0047	0.8760	-4.9505
13.0	0.200	0.209	0.0087	0.3794	-2.1444
19.2	0.200	0.231	0.0309	0.3972	-2.2452
25.3	0.200	0.031	0.2310	8.5327	-48.220
31.3	0.200	0.053	0.2524	9.2502	-52.275

The stability constants and Gibbs free energies of interaction between sodium Tungstate and Congo red dye in sodium perchlorate medium were calculated following equation given bellow explained in literature [23-39] and found to increase by increase in congo red dye concentration favoring more complexation [40-44]..

$$\Delta G = - 2.303\ RT \log \beta j$$

3.3. *Cyclic voltammogram for different concentrations of Na2WO4 in HCl (0.1) M:*

The cyclic voltammetry was performed for 0.1M HCl solutions and by adding different sodium tungstate (0.1M) using the glassy carbon electrode as the electrode that is in use (EGC). When the EGC is present, the waves have one primary reduction peak and are in the window range of 1.5V to -1.5V. at -0.1V. For further oxidation, two peaks are obtained here for the redox reaction for sodium tungstate in HCl medium. The main reduction peak at approximately - 0.3 V is very effective for the detection of tungstate ions in HCl medium. The Congo red dye effect on the redox reaction of tungstate ions was studied, and the data obtained are given in Figure (3) and Table (4).

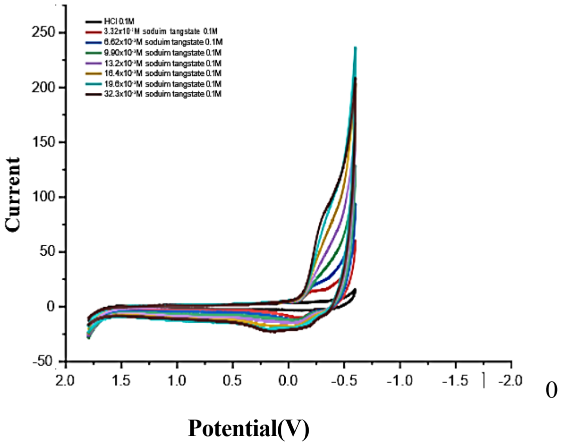

Fig. 3. Cyclic voltammogram for different concentrations of Na2WO4 in HCl (0.1) M at 0.1V/s and 302.85K.

Table 4 A:Effect of different concentrations of Na2WO4 by using EGC electrode at scan rate 0.1V/s and 302.85K.

[M] x10^3 mol.L^{-1}	Volt			Amp		Ip,a/Ip,c	Volt
	(-) Ep,a(V)	Ep,c(V)	ΔEp(V)	(-)Ip,ax10^6	Ip,cx10^6		E°
3.32	0.133	0.193	0.059	45.01	93.35	0.482	0.163
6.62	0.128	0.200	0.072	57.94	12.92	0.448	0.164
9.90	0.114	0.189	0.074	38.61	12.70	0.304	0.152
13.2	0.100	0.209	0.109	34.43	18.63	0.185	0.155
16.4	0.059	0.225	0.166	42.77	29.18	0.1465	0.142
19.6	0.055	0.281	0.225	44.26	53.50	0.0827	0.168
32.3	0.158	0.299	0.141	49.79	69.74	0.0713	0.228

Table 4 B: Continue

Dax10^5 cm^2.s^{-1}	Dc x10^5 cm^2.s^{-1}	Epc/2	Epa-Epc/2	αna	ksc x10^2 cm^2.s^{-1}	Γc x10^9 mol.cm^{-2}	(+)Qcx10^5 Columb	Γax10^9 mol.cm^{-2}	(-)Qax10^5 Columb
1.81	1.41	0.136	0.0570	0.850	4.63	8.040	4.87	3.877	2.35
1.17	6.79	0.160	0.0408	1.186	4.82	1.113	6.74	4.991	3.02
5.21	2.94	0.163	0.0259	1.866	4.18	1.094	6.63	3.325	2.02
3.50	3.58	0.161	0.0483	1.002	6.57	1.604	9.72	2.965	1.80
3.48	5.653	0.188	0.0379	1.278	2.78	2.513	1.52	3.68	2.23
3.01	1.328	0.217	0.0635	0.763	1.02	4.608	2.79	3.81	2.31
2.06	8.339	0.207	0.0917	0.528	1.35	6.006	3.64	4.29	2.60

3.4. *Cyclic voltammogram of presence of conco red with Na2WO4 in HCl (0.1) M:*

The reduction of congo red is represented by the observed reduction peak. This indicates that the congo red transforms into its leuco, or reduced, form by a reduction process. The leuco form of congo red dye, which is reduced, may not be stable under the experimental conditions because of the irreversibility of the reduction peak. This instability may cause additional reactions or changes in the leuco form. It appears that Na_2WO_4 does not significantly interact with conco red or its leuco form throughout the electrochemical process under study, based on the lack of effect it has on peak potential or peak current. This suggests that neither the reduction of congo red to its leuco form nor the next reactions are influenced by Na_2WO_4 Figure (4) and Table (5).The data given in Tables 4 A , 4B divided in three categories, diffusion, kinetic and adsorption parameters, are generally decreased by more adding congo red dye favouring mores solvation interaction between metal ions and dye ligand used.The **αna** and ksc kinetic parameters are decreased by more adding dye indicating decrease of the velocity of ions due to complexation [40-50].

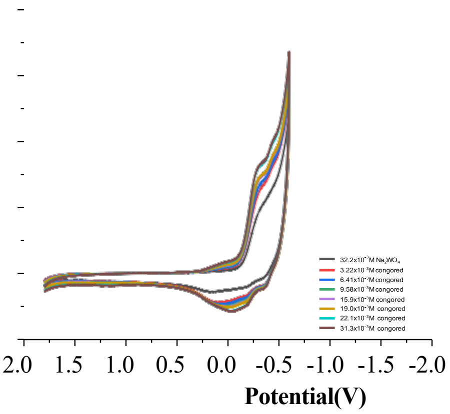

Fig. 4. Cyclic voltammogram for the interaction Na2WO4 with different concentrations of congo red using 0.1V/s and 302.85K.

The adsorption parameters, Γc, Γa, Qc, Qa in Table 5B are generally decreased by increase of Congo red dye supporting the complexation reaction between tungstate ion and the congo red dye.

Table 5 A :Effect of different concentrations of Congo red dye by using EGC electrode at scan rates (0.1 V/s and 302.85K)

[L]x10^3 mol.L^{-1}	**Volt**			**Amp**		**Ip,a/Ip,c**	**volt**
	(-)Ep,a(V)	Ep,c(V)	ΔEp(V)	(-)Ip,ax10^6	Ip,cx10^6		E°
3.22	0.073	0.309	0.381	10.03	12.14	0.082	0.118
6.41	0.068	0.301	0.369	12.17	12.71	0.095	0.116
9.58	0.022	0.315	0.337	21.89	13.66	0.160	0.146
15.9	0.007	0.325	0.317	15.53	14.04	0.110	0.166
19.0	0.029	0.320	0.291	21.55	14.11	0.152	0.174
22.1	0.052	0.303	0.251	20.43	15.83	0.129	0.177
31.3	0.047	0.314	0.267	26.84	15.79	0.169	0.180

Table 5 B: Continue.

Dcx10^5 cm^{-2}.s^{-1}	**Epc/2**	**Epa-Epc/2**	**αna**	**ksc x10^2 cm^{-2}.s^{-1}**	**Γc x10^9 mol.cm^{-2}**	**(+)Qcx10^5 columb**	**Γa x10^9 mol.cm^{-2}**	**(-)Qax10^5 columb**
3.275	-0.205	0.1041	0.464	2.19	1.045	6.34	8.64	5.23
1.366	-0.193	0.1083	0.447	1.77	1.094	6.63	1.05	6.35
2.714	-0.201	0.113	0.425	1.01	1.100	7.13	1.00	1.14
1.222	-0.203	0.121	0.400	6.92	1.209	7.33	1.34	8.11
1.214	-0.199	0.120	0.401	4.19	1.215	7.36	1.86	1.12
9.089	-0.194	0.1094	0.443	2.30	1.363	8.26	1.76	1.07
4.250	-0.195	0.118	0.408	2.22	1.360	8.24	2.31	1.40

Table 6 Stability constant for (Na_2WO_4+ congo red dye) interaction in HCl medium

[L]x10^3 mol.L^{-1}	**Metal** (Ep,a)M	**Complex** (Ep,a)C	**ΔE mv**	**log βj**	**ΔG (kJ /mol)**
3.22	0.228	0.118	0.11	3.68	-20.84
6.41	0.228	0.116	0.11	3.67	-20.78
9.58	0.228	0.146	0.082	2.60	-14.69
1.59	0.228	0.166	0.062	1.80	-10.21
1.90	0.228	0.174	0.054	1.45	-8.21

The different thermodynamic parameters are calculated here and found to be less in quantities than that using NaClO4 medium.The thermodynamic parameters, log βj and ΔG given in Table 6 are increased by more more increasing congo red dye concentration indicating complexation interaction(opposite Table 6 direction).

3.5. *Optimal supportive electrolyte's impact on free energy:*

From Figure (5), it is observed that the free energy for the NaClO4 signal is relatively large across the medium range tested. This could indicate a significant influence of NaClO4 on the electrochemical behavior or stability of the system under study. The use of HCl as another medium suggests a different set of conditions that could alter the electrochemical properties compared to $NaClO_4$. Stability of the system is crucial for accurate monitoring and interpretation of the voltammetric data. The system must remain stable for a predetermined amount of time to ensure reliable measurements of peak currents and potential variations. The variations observed in signal intensity and potential peaks when changing the media (NaClO4 and HCl) indicate that these media have differing effects on the electrochemical behavior of Na2WO4, especially in the presence or absence of Congo red dye.

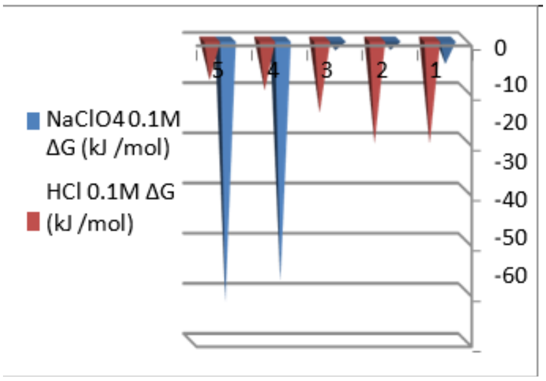

Fig. 5. Effect of the ideal supporting electrolyte for free energies with 0.1M NaClO4 and 0.1M HCl

4. Conclusions

The interaction solvation parameters, particularly the adsorption parameters such as the anodic quantity of electricity (Γa), cathodic surface coverage (Γc), and anodic surface coverage, indicate that strong interactions significantly influenced the cathodic quantity of electricity (Qa) and Gibbs free energies of interaction. Specifically, the interaction of Na2WO4 with Congo red dye in 0.1M NaClO4 exhibited notable strength, suggesting that the electrochemical environment facilitated these interactions more effectively than other media, such as 0.1M HCl**.** Moreover, all stability constants and Gibbs free energy values associated with these interactions were indicative of strong electrostatic interactions. This suggests that the forces driving the interactions between Na_2WO_4 and Congo red dye are primarily electrostatic in nature, which enhances the overall stability and effectiveness of the adsorption process in the $NaClO_4$ medium. The findings highlight the importance of the solvation environment in modulating the interaction parameters, with $NaClO_4$ proving to be particularly effective in promoting strong electrostatic interactions.

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