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Determination of Formation Constant and Thermodynamic Parameters for Interaction between Vanadyl Sulfate and Gliclazide in Aqueous – Organic Solvent at Various Temperatures

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Authors' contributions

This work was carried out in collaboration between all authors. Author EAG designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors AN and MMR managed the analyses of the study. Author MMR managed the literature searches. All authors read and approved the final manuscript.

Article Information

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Short Research Article

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ABSTRACT

THURSDAY

Our work aims to measure the conductivity of vanadyl complexes in an aqueous-organic medium at various temperatures ranging from (298.15, 303.15, 308.15 and 313.15 K). The experimental data of *Λm* and *Λo* were dissected in place of the calculation of formation constants for every form of the stoichiometric complexes. The formation constants (K_f) for the metal complexes were studied for every form of complexes obtained 1:1 and 1:2 metal to ligand molar ratio (M:L) via using formation equation. It is observed that the limiting equivalent conductance increased with the increment in temperature. Thermodynamic parameters (i.e*., ΔG⁰ f*, *ΔH0 f*, *ΔS⁰ ^f*) are estimated from the temperature dependence of the ion formation constant. Vanadyl complexes show formation within experimentally various temperatures which are supported by the negative value of *ΔG⁰ ^f*. The positive values of *ΔS⁰ ^f* in most cases and positive values of *ΔH⁰ ^f* indicate that the formation process occurred spontaneously as well as endothermic and vice versa at all respective temperatures.

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1. INTRODUCTION

Conductance measurement is one of the most accurate and widely used physical methods for investigation of the electrolyte of solutions [1,2]. Conductivity measurements are widely used in industry. Some studies have used the electrolytic conductance measurements for studying the interaction of some ligands with some metal cations and determination of the formation constants of the formed complexes. Some physicochemical techniques such as spectrophotometer and polarography have been used [3–5] to study the complex formation between GL and vanadyl sulfate in the solutions. The conductometric technique is a sensitive and inexpensive method with a simple experimental arrangement for such investigations. Conductometric analysis can be used for many titration procedures where ionic species are involved. As the conductance of a solution relates to the total ionic content, it can be used to follow reactions that result in a change in this quantity. Conductometry is an electrochemical technique, which provides us with most precise data for ions in the solution [6]. There is an increasing interest in the study of complexation reactions in different solvent systems and their interpretation in terms of solute's preferential salvation by one of the mixed solvent components [7–9].

Dimethylformamide (DMF) acts as a strong solvent which characteristic by the most benefited in fiber and film production and also in a coating and adhesive formulations. DMF used as a solvent due to it is the most economic solvent when comparing with the cheap solvents due to its solving more solid substances in practical operating viscosities. DMF is especially an efficient solvent with vinyl polymers, polyacrylonitrile, epoxy resins, polyamides, and urethanes. Due to its high purity and selective solvent characteristic, DMF is used for instance in removing or gaining acetylene, and removing butadiene from hydrocarbons processes.

Vanadyl sulfate is an inorganic compound of vanadium(V). This highly hygroscopic substance blue solid is one of the maximum common sources of V in the lab, reflecting its high stability. It features the vanadyl ion, $VO²⁺$, which has been called the "most stable diatomic ion" [10] It is a major commercial source of V and also an

intermediate in the extraction of V from petroleum residues. It is a component of some experimental drugs and food supplements. VOSO4 exhibits insulin-like effects [11].

Gliclazide (GL) is an oral antihyperglycemic agent, it used for medicament of non-insulin dependent diabetes mellitus which belongs to the sulfonylurea class of insulin secretagogues. It works by stimulating β cells of the pancreas to release insulin [12].

In this work, the formation constants and stoichiometry of the complexation were elucidated by conductometric titration of VO^{2+} solutions with GL solution. The conductometric method for the study of $(VO-GL)^{2+}$ complexes in various $H₂O/DMF$ mixtures and temperatures were used to investigate the effect of solvent properties on the stabilities of the resulting.

2. EXPERIMENTAL

2.1 Materials

Vanadyl sulfate $(VOSO₄.3H₂O$, 99.5%), Gliclazide and DMF (99.5%) were supplied from Sigma-Aldrich company, which used without further refining. Deionized water which has a specific conductivity = $0.07 \mu S \text{ cm}^{-1}$ at 25°C was furthermore used for binary solvents preparation [13-16].

Specific conductance measurements of the salt solutions that previously prepared were
measured by using LF 191 (Germany) measured by using LF conductivity meter with accuracy ±0.01% of a cell constant value 1±10% cm−1 . The cell constant was determined with KCl solutions [17-21]. MLW 3230 ultra-thermostat was connected to the Conductivity Bridge to give constant temperature $(\pm 0.006^{\circ}C)$.

A solution of VOSO₄ (1x10⁻³ mol·dm⁻³) was put into a titration cell. Thermostated at used the temperature and also measured the conductance of the solution [22-25]. The ligand gliclazide $(1x10⁻⁴$ mol.dm⁻³) was transported step by step to the titration cell using a recalibrated micropipette. After each transfer measured the conductance of the solution. The addition of the gliclazide solution was continued until the total concentration of the gliclazide was higher four times approximately than the Vanadyl ions. The

conductance of the solution was measured by titration of gliclazide as a ligand with Vanadyl sulfate as a metal salt. The molar conductance of the complexes ML and the formation constant of the complexes (K_f) were estimated by computer fitting to the molar conductance mole ratio data. The temperatures used are 298.15, 303.15, 308.15 and 313.15 K.

3. RESULTS AND DISCUSSION

3.1 Determination of Molar Conductance

The specific conductance of solutions understudy of the salt with a concentration range of (1×10^{-3}) - 7.1 x 10⁻⁴ mol.L⁻¹) in binary mixed solvents with the alcohol mass fractions of 0, 30 and 60% of DMF-water at various temperatures (298.15, 303.15, 308.15 and 313.15 K) was obtained. The molar conductance ($Λ_m$) for VOSO₄ solutions in the presence of gliclazide in used solvents at various temperatures was determined by using equation (1) and summarized in Table 1.

$$
A_m = \frac{(K_s - K_{solv}) \cdot K_{cell} \cdot 1000}{C} \tag{1}
$$

Where $(K_s, \mu S \text{ cm}^{-1})$ is the obtained specific conductance of the solution under study and K_{solv} is the specific conductance of used solvent, K_{cell} is the cell constant and C is the molar concentration of the metal salt $(VOSO₄)$ solution.

From Table 1, when comparing the obtained data for $VOSO₄$ in the presence of gliclazide, we observed that the value of molar conductance (*Λm*) was inversely proportional with increasing the fraction of the organic-aqueous solvent as shown in Fig.2 so has the following direction 60% $<$ 30% $<$ 0% [26]. This is related to solventsolvent interaction (the formation of

intramolecular and intermolecular hydrogen bonding) between DMF and water which create impediment to the mobility of ions, reduce the dissociation and solvation of $VOSO₄$ molecules, and also the electrostatic force between vanadyl sulfate and gliclazide play an important role in this decreasing the value of molar conductance. Also, we observed that the increase in values of limiting molar conductance for $VOSO₄$ in used solvents with rising temperature as shown in Fig. 1 this is because of increasing of kinetic energies of ions in solution.

3.2 Determination of Formation Constant

By drawing the relation between molar conductance (*Λm*) for VOSO4 in presence of gliclazide and its molar ratio of [M]/[L] concentrations at various temperatures, the different lines were obtained with two breaks which indicates formation of two complexes which have different stoichiometry as the following 1:2 and 1:1 (M:L) complexes ,as done in previous works [27-28]. See Figs. 3-5.

By using equation (2) [29-32] to calculate formation constants (K_f) for VOSO₄ complexes for every form of complexes 1:1 and 1:2 (M:L) with gliclazide.

$$
K_{f} = \frac{\Lambda_{m} - \Lambda_{obs}}{(\Lambda_{obs} - \Lambda_{ML})[L]}
$$
 (2)

Where *Λobs* is the molar conductance of solution during titration, *Λm* is the limiting molar conductance of the VOSO₄ alone, Λ_{ML} is the molar conductance of the complex and [L] is the ligand concentration. The obtained values of the formation constant (K_f) for all studied systems were summarized in Tables 2 and 3.

Fig. 1. Conductometric titration curves for VOSO4 in the presence of GL in pure H2O at various temperatures

0.2 0.4 0.6 0.8 1.0 1.2 1.4 1.6 1.8 2.0 2.2

[M]/[L]

Solvent	$C[M]/[L]$ (mol. d m ⁻³)	$\overline{\Lambda(}S \text{ cm}^2 \text{ mol}^1)$			
		298.15	303.15	308.15	313.15
0%(DMF-H ₂ O)	0.3	2300.33	2506.33	2643.66	2884
	0.4	1898	2002	2106	2236
	0.5	1617	1722	1785	1890
	0.6	1431	1501.66	1590	1678.33
	0.7	1299.28	1375.71	1467.42	1528.57
	0.8	1201.5	1269	1350	1444.5
	0.9	1150.55	1199	1283.77	1356.44
	$\mathbf{1}$	1089	1161.6	1210	1287
	1.1	1059.54	1110	1160.45	1241.18
	1.2	1017.33	1073.33	1110.66	1176
	1.3	973.54	1025.69	1077.84	1138.69
	1.4	944.57	985.28	1034.14	1107.42
	1.5	920	958.33	1019.66	1081
	1.6	841	935.25	993.25	1044
	1.7	825.88	922.23	956.65	1025.47
	1.8	812.88	891.55	944	1003
30%(DMFH ₂ O)	0.3	1078.06	1091.8	1133	1201.66
	0.4	832	858	910	988
	0.5	714	735	777	840
	0.6	636	671.33	706.66	742
	0.7	580.857	596.14	642	672.57
	0.8	553.5	580.5	621	648
	0.9	520.777	545	581.33	617.66
	$\mathbf{1}$	495	525.25	547.25	583
	1.1	484.363	504.54	524.72	565.09
	1.2	457.333	485.33	513.33	550.66
	1.3	452	469.38	495.46	530.23
	1.4	439.714	464.14	488.57	513
	1.5	429.333	452.33	475.33	498.33
	1.6	420.5	442.25	471.25	493
	1.7	412.941	426.70	468	474.88
	1.8	406.444	419.55	458.88	465.44
60%(DMFH ₂ O)	0.3	789.66	930.77	1063.64	1030
	0.4	598	728	858	858
	0.5	483	588	693	693
	0.6	406.33	530	618.33	583
	0.f	397.42	458.57	535	519.71
	0.8	351	405	472.5	459
	0.9	314.88	363.33	423.88	411.77
	$\mathbf{1}$	286	336.05	385	378.18
	1.1	262.36	343.09	383.45	373.36
	1.2	270.66	317.33	354.66	345.33
	1.3	252.07	295.53	330.31	339
	1.4	236.14	276.85	309.42	317.57
	1.5	222.33	260.66	291.33	299
	1.6	210.25	268.25	290	297.25
	1.7	220.23	254.64	275.29	282.17
	1.8	209.77	242.55	262.22	268.77

Table 1. The molar conductance (Λm±0.07, S cm2 mol−1) for VOSO4 in the presence of gliclazide at various temperatures in the used solvents

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Fig. 3. Conductometric titration curves for VOSO4 to GL in water at 298.15K

Fig. 4. Conductometric titration curves for VOSO₄ to GL in 30% (DMF-H₂O) at 303.15K

Fig. 5. Conductometric titration curves for VOSO₄ to GL in 60% (DMF-H₂O) at 313.15K

Solvent	T/K	$^{\circ}$ (mol. d m $\overline{^{3})}$ x10 $^{\circ}$ [L]t $x10^\circ$	[M]tx10 ⁵ (mol. d m ⁻³)x10 ⁵	Λ _o (S cm ² .mol ⁻¹)	$\Lambda_{\text{obs.}}$ (S cm ² .mol ⁻¹)	$log K_f$ (mol/dm ³)
0% (DMF- H_2O)	298.15	9.524	4.761	2300	1617	3.988
	303.15			2506.33	1722	4.055
	308.15			2660.83	1785	4.127
	313.15			2884	1890	4.199
30% (DMF-H ₂ O)	298.15	9.524	4.761	1078	714	4.10
	303.15			1091.8	735	4.05
	308.15			1133	777	4.00
	313.15			1201.66	840	3.95
60% (DMF-H ₂ O)	298.15	9.524	4.761	789.67	483	4.428
	303.15			930.6	588	4.316
	308.15			1063.65	693	4.217
	313.15			1030	693	4.118

Table 2. Limiting molar conductance (Λ。)(±0.05S cm 2 **.mol** 1 **), formation constant (K_t) (±0.02 mol/dm** 3 **), for VOSO₄ to gliclazide (GL) complex (1:2) formation at various temperatures in used solvents**

Table 3. Limiting molar conductance (Λ $_{\rm o}$ **) (±0.05 S cm** 2 **.mol** 1 **), formation constant (K** $_{\rm f}$ **) (±0.02 mol/dm** 3 **), for VOSO** $_4$ **to gliclazide (GL) complex (1:1) formation at various temperatures in used solvents**

From Tables 2 and 3 we observed that the formation constant for $VOSO₄$ complexes with stoichiometry $(1:1)$ in 30%, 60% (DMF-H₂O) and (1:2) stoichiometry in 0% (DMF-H₂O) increases with temperature increase, so it shows that the complexation is thermopile reaction which showed an exothermic formation process. The formation process of complexes mainly dependence on temperature which showed that the relation between complexation and desolvation of ions by increasing temperature, the desolvation of ions take a great place which caused of the ions will be nearer than before by electrostatic force between ions, so the formation of complexes increased [33]. But when comparing the values of the formation constant for $VOSO₄$ complexes with stoichiometry (1:2) in 30% , 60% (DMF-H₂O) and (1:1) stoichiometry in 0% (DMF- H_2O) decreases with temperature increase which showed an endothermic formation process.

3.3 Determination of Thermodynamics Parameter

The Gibbs free energy of formation for every stoichiometry complex $(AG^{\mathcal{O}}_t)$

Fig. 6. log K_f against 1/T for VOSO₄ to GL in **water.**

was determined by applying equation (3) [34-38]:

$$
\Delta G_f^0 = -2.303 RT \log K_f \tag{3}
$$

By drawing the relation between *1/T* and log *Kf*, for all complexes strait lines were obtained as shown in Figs. 6, 7 and 8) as explains in previous works [35-40].

The change in the standard enthalpy (ΔH_{f}^{0}) for the complexation reaction was determined in the usual manner from the slope of the *Van't Hoff* plot which equal *(-ΔH⁰ ^f /2.303R*). The change in entropy (*ΔS0 ^f*) for VOSO4 stoichiometric complexes were determined [31] for every form of complexes (1:2) and (1:1) (M: L) by applying Gibbs-Helmholtz equation (4) [41-45]:

$$
\Delta G_f^0 = \Delta H_f^0 - T \Delta S_f^0 \tag{4}
$$

The thermodynamic parameters (*ΔH0 f, ΔS0 f, and ΔG⁰ ^f*) for the complexation between vanadyl sulfate and gliclazide are summarized in Tables 4, 5.

Fig. 7. log K_f against 1/T for VOSO₄ to GL in **30% (DMF-H2O)**

Fig. 8. log K_f against 1/T for $VOSO₄$ to GL in 60% (DMF-H₂O)

Solvent	T/K	$\Delta G^{\mathrm{U}}_{\mathrm{f}}$ (kJ mol ⁻¹)	$\Delta H_{\text{f}}^{\text{0}}$ (kJ.mol ⁻¹)	ΔS_f (J mol ⁻¹ K ⁻¹)
0% (DMF-H ₂ O)	298.15	-22.77	25.11	160.61
	303.15	-23.54	25.11	154.52
	308.15	-24.35	25.11	160.54
	313.15	-25.18	25.11	160.60
30% (DMF-H ₂ O)	298.15	-23.78	-19.23	15.27
	303.15	-23.89	-19.23	15.38
	308.15	-23.96	-19.23	15.36
	313.15	-24.01	-19.23	15.27
60% (DMF-H ₂ O)	298.15	-25.27	-36.69	-38.27
	303.15	-25.05	-36.69	-38.38
	308.15	-24.88	-36.69	-38.31
	313.15	-24.69	-36.69	-38.30

Table 4. Gibbs free energies (ΔG 0 **_f) Enthalpy change (ΔH** 0 **_f) and entropy change (ΔS** 0 **_f) for VOSO4 formed complexes (1:2 M/L) with GL at various temperatures in used solvents**

Table 5. Gibbs free energies (ΔG 0 **_f) Enthalpy change (ΔH** 0 **_f) and entropy change (ΔS** 0 **_f) for VOSO4 formed complexes (1:1 M/L) with GL at various temperatures in used solvents**

Generally; it was noticed that the thermodynamic parameters were affected by the temperature as the temperature increase the negativity of free energies (*ΔG⁰ ^f*) increase as well, this shows that the systems under study are spontaneous; this clearly shows that the complex formation process prefers a lower value of the dielectric constant medium.

Enthalpy (*ΔH° ^f*) has positive values in pure water with stoichiometric (1:2), 30% and 60% DMF-H₂O with stoichiometric (1:1) this clearly shows that the formation reactions in these cases are endothermic, in the meantime other cases with negative values are exothermic reactions.

In all cases, the entropy (*∆S° ^f*) has positive values so the spontaneity of the reaction is more favored for these complexation processes. As the positivity increase the degree of disorderliness increase as well. Except in case of 60% (DMF-H2O) 1:2 stoichiometric we observed

that the entropy has negative value. In natural systems entropy known as an increase of randomness or disorder, and negative entropy means an increase of organization or orderliness. Negative entropy is also known as negentropy. Individual systems can experience negative entropy, but overall, natural processes in the universe trend toward entropy.

Thermodynamic parameters changes for the $complexation$ process in $(DMF-H₂O)$ were determined by the solvent composition. By applying Raoult's law for vapor pressure depression of binary systems, it was noticed a great deviation from ideal behavior of the binary systems, this was due to interaction between solvent's molecules, because DMF is a dipolar solvent which enables the formation of strong hydrogen bond with water; by acquiring its hydrogen atoms especially when totally miscible with water, Doubly bound oxygen is usually a good hydrogen acceptor. When water is mixed with strong hydrogen-acceptors.

4. CONCLUSIONS

The results obtained in this work showed that the composition of the binary solution and solvent properties are very important since the stability depends on the solvent-solvent interaction nature, which takes a great place in the solution. As the temperature increases the formation constant increase as well, and also with the proportion of DMF increases. Even the exceptional manner of 60% (DMF-H₂O) mixed solvent was studied. The more negative values of *ΔG° ^f* showed that the formation reaction is spontaneous in nature. The positive charge of *ΔS° ^f* is indicating that the entropy is the driving force for the formation reaction except in case of 60% (DMF-H2O) in 1:2 stoichiometric has a negative charge. The positive charge of *ΔH° ^f* is showed that the formation process is endothermic. The negative charge of the enthalpy change (*ΔH° ^f*) is showed that the formation process is exothermic. Conductometric measurements revealed the 1:1 [LM] stoichiometry complexes more expected than 2:1 complex $[L₂M]$.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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