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Dependence on Ionic Strength of Formation Constants, Protonation, and Complexation of Nitrilotriacetic Acid with Tungsten(VI) in Sodium Perchlorate Aqueous Solution

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By use of potentiometric and spectrophotometric techniques, the complexation of tungsten(VI) with nitrilotriacetic acid (NTA) has been carried out in aqueous solution for pH = 7.5 at 25 °C and different ionic strengths ranging from (0.1 to 1.0) mol dm^{-3} ($NaClO_4$). The composition of the complex was determined by the continuous variations method. It was shown that tungsten(VI) forms a mononuclear complex with NTA of the type (WO_3L^{3-}) at pH = 7.5. The dependence of the protonation of the aforementioned ligand and the stability constant of the complex on ionic strength is described by a Debye−Huckel-type equation. Finally the results have been compared with the previously reported data.

Introduction

Desulfovibrio gigas formate dehydrogenase is the first representative of a tungsten-containing enzyme from a mesophile that has been structurally characterized.[1] It is a heterodimer of 110 and 24 kDa subunits. The existence of a universal pterin dithiolene cofactor ligand for the molybdenum and tungsten oxotransferases supports a biological significance of the fundamental chemistry of mono- and bis(dithiolene) complexes of these elements.[2] Tungsten(VI) also forms complexes with tryptophan[3] and porphyrin.[4] By pursuing our systematic study on complexation of molybdenum (VI) with nitrilotriacetic acid (NTA), we thought it interesting to investigate the protonation and complexation of NTA with tungsten(VI) at different ionic strengths of sodium perchlorate.

Considerable studies have been performed on the ionic strength dependence of the formation constants of some weak complexes of alkali and alkaline earth metals with some organic and inorganic ligands,[5−13] and little work has been carried for more stable complexes of α-amino acids with transition-metal ions,[14−17] but according to literature, no work has been reported on the ionic strength dependence of formation constants, protonation, and complexation of tungsten(VI) with NTA.

Marcu[18] studied the radiochromatographic and electroradiochromatographic of sodium tungstate solutions under the action of NTA. Studies on the structural and bonding characteristics of various Mo(VI)−aminopolycarboxylic acid complexes has led to the evaluation of stability constants from proton nuclear magnetic resonance (NMR) data.[19,20] On the basis of these studies, Kula[21] determined the stability constants of W(VI)−NTA complexes by potentiometric techniques. Collin[22] reported the stability constant of this system at 25 °C and at an ionic strength of 0.5 mol dm^{-3} $NaClO_4$. Zare[23] studied this system and evaluated its stability constant at 25 °C and at an ionic strength of 3 mol dm^{-3} $NaClO_4$ using the potentiometric technique. Anderegg[24] has made a critical survey of stability constants of NTA complexes.

In the present work, we have studied the complexes of W(VI) with NTA in an ionic strength range of (0.1 to 1.0) mol dm^{-3} sodium perchlorate at 25 °C. A simple Debye−Huckel-type equation was established for the dependence of formation constants on ionic strength. This equation makes it possible to estimate a stability constant at a fixed ionic strength when its value is known at another ionic media in the range ($0.1 < I < 1.0$) mol dm^{-3}.

Experimental Section

Reagents. Sodium perchlorate, perchloric acid, sodium hydroxide, sodium tungstate, and NTA were obtained from E. Merck as analytical reagent grade materials and were used without further purification. Dilute perchloric acid solution was standardized against $KHCO_3$. In all experiments, double-distilled water with specific conductance equal to (1.3 ± 0.1) $\mu\Omega^{-1}$ cm^{-1} have been used.

Measurements

A Horiba pH meter, D-14, was used for pH measurements. The pH meter has a sensitivity of 0.01 units.The hydrogen ion concentration was measured with a Horiba combination electrode, model S8720. A 0.01 mol dm^{-3} perchloric acid solution containing 0.09 mol dm^{-3} sodium perchlorate (for adjusting the ionic strength to 0.1 mol dm^{-3}) was employed as a standard solution of hydrogen ion concentration. The same procedure was performed for the other ionic strengths.[23] The calibration has been done for the whole pH (pH = $-\log[H^+]$) range used. Spectrophotometric measurements were performed on a UV−vis Shimadzu 2101 spectrophotometer with an Acer Mate 486 SX/250 computer using thermostated, matched 10-mm quartz cells.

For each experiment two solutions of W(VI) + NTA were prepared with the same concentration, but the ionic strength of the first was maintained with sodium perchlorate and that of the second with sodium hydroxide or

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Table 1. Potentiometric Data for the Titration of NTA with Sodium Hydroxide at $I = 0.1$ mol dm^{-3} of $NaClO_4$ and 25 °C

V/mL	pH	V/mL	pH	V/mL	pH	V/mL	pH
0.00	2.49	1.64	3.69	2.00	8.86	3.66	11.04
0.01	2.50	1.67	3.79	2.02	8.93	3.72	11.11
0.02	2.50	1.70	3.93	2.04	8.98	3.78	11.17
0.04	2.51	1.72	4.06	2.06	9.04	3.85	11.23
0.06	2.51	1.74	4.20	2.08	9.09	3.95	11.32
0.09	2.52	1.75	4.32	2.10	9.13	4.05	11.39
0.12	2.53	1.76	4.41	2.12	9.18	4.15	11.45
0.16	2.54	1.77	4.51	2.15	9.25	4.25	11.49
0.20	2.55	1.78	4.68	2.18	9.32	4.40	11.57
0.30	2.58	1.79	5.03	2.21	9.37	4.60	11.65
0.40	2.62	1.80	5.44	2.25	9.45	4.80	11.71
0.45	2.64	1.81	5.78	2.29	9.52	5.10	11.79
0.50	2.65	1.82	6.36	2.33	9.60	5.40	11.85
0.60	2.70	1.83	6.84	2.35	9.63	5.80	11.93
0.70	2.74	1.84	7.20	2.40	9.72	6.20	11.99
0.80	2.79	1.85	7.66	2.45	9.81	6.70	12.04
0.90	2.84	1.86	7.81	2.50	9.89	7.20	12.09
1.00	2.90	1.87	7.96	3.10	10.00	7.70	12.14
1.10	2.96	1.88	8.10	3.16	10.10	8.20	12.18
1.20	3.04	1.89	8.23	3.22	10.22	9.00	12.24
1.30	3.13	1.90	8.34	3.28	10.32	10.00	12.31
1.35	3.18	1.91	8.37	3.34	10.44	11.00	12.35
1.40	3.24	1.93	8.52	3.40	10.55	12.00	12.44
1.45	3.30	1.95	8.65	3.46	10.66	14.00	12.50
1.50	3.38	1.96	8.70	3.52	10.77	16.00	12.55
1.55	3.47	1.97	8.74	3.58	10.87		
1.60	3.57	1.98	8.80	3.64	10.96		

perchloric acid. The pH of the first solution was adjusted with the second one. The second solution consists of metal + ligand + NaOH for increasing pH, while for decreasing pH, the second one consists of metal + ligand + $HClO_4$. The absorbance of the first solution was measured after adjusting the pH. In all cases, the procedure was repeated at least three times and the resulting average values and corresponding standard deviations are shown in the text and Tables.

Results and Discussion

The dissociation equilibria of NTA have been studied in different kinds of background electrolytes but there are no reports about the ionic strength dependence of the dissociation constants of NTA.[25] The following equilibria were studied

$$H_3L \rightleftarrows H^+ + H_2L^- \qquad K_1 = [H^+][H_2L^-]/[H_3L] \quad (1)$$

$$H_2L^- \rightleftarrows H^+ + HL^{2-} \qquad K_2 = [H^+][HL^{2-}]/[H_2L^-] \quad (2)$$

$$HL^{2-} \rightleftarrows H^+ + L^{3-} \qquad K_3 = [H^+][L^{3-}]/[HL^{2-}] \quad (3)$$

Where L^{3-} represents the fully dissociated NTA anion. Three titrations have been done for each ionic strength, and 106 points have been used for calculations at each ionic strength.The dissociation constants K_1, K_2, and K_3 have been determined using potentiometric techniques and calculated using Microsoft Excel 2000 program.[25,26] Potentiometric data are listed in Table 1. Dissociation constants are given in Table 2 together with the values reported in the literature, which are in good agreement with those reported before.

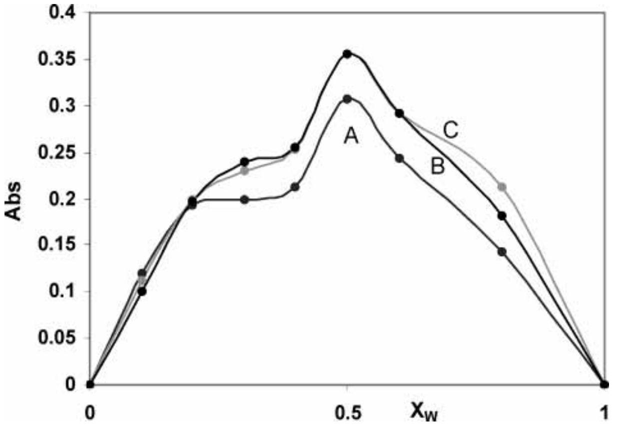

Figure 1. Continuous-variation plots of the absorbances of WO_3L^{3-}, abs, vs the mole fraction of W(VI), X_W, at 25 °C, an ionic strength of 0.1 mol dm^{-3} $NaClO_4$, and different wavelengths: A, 260 nm; B, 265 nm; and C, 270 nm.

Complexation of Tungsten(VI) with NTA

By use of the continuous variations method, the absorbances of solutions of W(VI) and NTA of total concentration 0.006 mol dm^{-3} in the UV range (260−270 nm) at a constant pH of 7.5 were determined. When solutions of tungstates are made weakly acidic, polymeric anions are formed, but from more strongly acid solutions, substances often called tungstic acid are obtained.[27] The behavior of the tungstate systems is similar to that of the molybdate systems. Again the degree of aggregation in solution increases as the pH is lowered, and numerous tungstates have been crystallized from the solutions at different pHs.[27] Tungsten(VI) and the complex both have absorbances at the specified wavelengths, so the absorbance of the tungsten should be eliminated from the total absorbance (A_{obs}). The observed absorbances were corrected from eq 4 and are summarized in Table 3 and plotted in Figure 1

$$A_c = A_{obs} - \epsilon_0[W] \qquad (4)$$

A_c, A_{obs}, and ϵ_0 are the absorbance of the complex, the observed absorbance, and the molar absorptivity of W, respectively. ϵ_0 values were calculated at the mole fraction of W equal to 1 and are shown in Table 4. In Figure 1, a maximum at a mole fraction of W equal to 0.5 was obtained, indicating a 1:1 complex. The molar absorptivity of the complex, ϵ_1, was calculated from the linear part of the aforementioned plot at low mole fraction of W, where essentially all the metal ions were in the form of a complex, and is listed in Table 4. At the maximum point of the plot,

Table 2. Dissociation Constants K_3, K_2, and K_1 of NTA at Different Ionic Strengths, I, of $NaClO_4$

I/mol dm^{-3}	log K_3	log K_2	log K_1	experimental conditions	ref
0.1	1.98 ± 0.02	2.92 ± 0.05	10.00 ± 0.02		this work
0.3	1.83 ± 0.04	2.84 ± 0.03	9.85 ± 0.01		this work
0.5	1.76 ± 0.01	2.79 ± 0.02	9.70 ± 0.03		this work
0.7	1.63 ± 0.02	2.61 ± 0.01	9.55 ± 0.02		this work
1.0	1.55 ± 0.02	2.59 ± 0.04	9.25 ± 0.02		this work
			9.81 ± 0.10	$I = 0.15$ M, $T = 25$ °C	21
	2.05 ± 0.05	2.63 ± 0.02	9.17 ± 0.04	$I = 3$ M $NaClO_4$, $T = 25$ °C	23
	1.65	2.94	10.33	$T = 20$ °C	29

Table 3. Continuous-Variation Data for the W(VI) + NTA System at pH 7.5, an Ionic Strength of 0.1 M NaClO$_4$, and Different Wavelengths

mole fraction of W(VI)	A^a 260 nm	265 nm	270 nm
0.00	0.000	0.000	0.000
0.10	0.120	0.112	0.100
0.20	0.194	0.200	0.198
0.30	0.200	0.230	0.240
0.40	0.213	0.254	0.255
0.50	0.307	0.356	0.356
0.60	0.243	0.292	0.292
0.80	0.143	0.212	0.182
1.00	0.000	0.000	0.000

[a] The corrected absorbance of [W(VI)] + [NTA] = 0.006 mol dm^{-3}.

Table 4. Molar Absorptivities of W(VI), ϵ_0, and WO$_3$L^{3-}, ϵ_1, at pH 7.5, Different Wavelengths, and Various Ionic Strengths, *I*, of NaClO$_4$

	ϵ_0			ϵ_1		
I/mol dm^{-3}	260 nm	265 nm	270 nm	260 nm	265 nm	270 nm
0.1	660.8	486.6	486.6	1116.6	1116.6	1116.6
0.3	527.5	365.0	246.1	1221.6	1268.3	1250.0
0.5	505.8	345.0	245.8	1533.3	1583.3	1716.6
0.7	478.3	320.0	224.1	716.6	750.0	766.6
1.0	522.5	355.0	251.6	2316.6	2350.0	2333.3

the concentration of the complex is

$$[\mathrm{C}] = A_c/\epsilon_1 \tag{5}$$

It is now certain that the WO$_4^{2-}$ ion is tetrahedral in aqueous solution.[27] In the usual potentiometric method for evaluating metal−ligand stability constants, the competition between the metal ion and the hydrogen ion for the ligand is studied, and the pH region of interest is from about 1 to 5.[21] In the W(VI) systems, however, the complication of metal polymerization is introduced in acidic solutions. Because the polymerization equilibria are not well understood, this pH region is not useful for stability determinations. In more alkaline solutions, on the other hand, a pH-dependent process involving the competition between tungstate formation and metal−ligand complexation, i.e., a competition between OH$^-$ and ligand for the metal ion, can be utilized. This process was determined from the NMR studies to be important from about pH 6 to 9 and can be represented by[21]

$$\mathrm{MO_3L^{x-} + OH^- \rightleftarrows MO_4^{2-} + HL^{1-x}} \tag{6}$$

where M represents W and L represents the aminopolycarboxylic acid ligand. In the pH region above 6, no evidence was found for any Mo species containing fewer than three oxygen atoms, e.g., MoO$_2^{2+}$, as has been proposed for other systems.[28] The molybdenum-coordinating species in all the aminopolycarboxylic acid systems above pH 6 is MoO$_3$, and by analogy, we have assumed that the corresponding coordinating unit in the tungsten systems is WO$_3$.

By use of the potentiometric technique, results obtained for simple one to one metal−ligand chelates are more precise than for higher complexes, such as those formed with EDTA. In fact, for the multicomplex systems, the NMR data are probably more reliable than the potentiometric data as indicated by the range of calculated values for the EDTA systems.[21] A comparison of the formation constants shows that within experimental error there is essentially no difference between the stabilities of the corresponding Mo(VI) and W(VI) chelates. This does not seem too surprising in view of the similarities of the two ions and their nearly equal ionic radii.[21] However, NMR studies show that tungstene chelates are significantly more labile with respect to individual metal−ligand bonds than are the molybdenum chelates. The slowness with which pH equilibrium was attained in the NTA chelates may be explained by the higher negative charge of this complex compared to the other complexes and the subsequently slower reaction with OH$^-$

$$\mathrm{MO_3NTA^{3-} + OH^- \rightarrow MO_4^{2-} + HNTA^{2-}} \tag{7}$$

Table 5. Average Values of β_{112} at pH 7.5 and Different Ionic Strengths for the Complexation of Tungsten(VI) with NTA, *T* = 25 °C

I/mol dm^{-3}	log β_{112}	experimental conditions	ref
0.1	19.00 ± 0.30		this work
0.3	19.37 ± 0.10		this work
0.5	19.65 ± 0.25		this work
0.7	20.10 ± 0.10		this work
1.0	19.40 ± 0.20		this work
	18.86 ± 0.05	*I* = 0.15 M, *T* = 25 °C	21
	19.10 ± 0.2	*I* = 1.0−2.5 M, *T* = 35 °C	21
	17.75	*I* = 0.5 M NaClO$_4$, *T* = 25 °C	22
	19.03 ± 0.15	*I* = 3 M NaClO$_4$, *T* = 25 °C	23

For the NTA ligand system, NMR studies also indicate that only one metal−ligand species exists above pH 6, MO$_3$L^{x-}.[21] Thus, W(VI) will bond with this tridentate ligand as a 1:1 complex

$$\mathrm{WO_4^{2-} + L^{3-} + 2H^+ \rightleftarrows WO_3L^{3-} + H_2O} \tag{8}$$

with the stability constant, β_{112}, as

$$\beta_{112} = [\mathrm{WO_3L^{3-}}]/[\mathrm{WO_4^{2-}}][\mathrm{L^{3-}}][\mathrm{H^+}]^2 \tag{9}$$

The following equations are valid for the total concentration of tungsten (C_M) and the total concentration of the ligand (C_L) at the maximum point on the plot of Figure 1

$$C_\mathrm{M} = [\mathrm{W}] + [\mathrm{C}] \tag{10}$$

$$C_\mathrm{L} = [\mathrm{L}] + [\mathrm{C}] \tag{11}$$

[C] is the concentration of the complex. By substitution of eqs 5, 10, and 11 in eq 9, we can calculate the values of β_{112} according to reaction 8. Stability constants have been calculated by the combination of spectrophotometric and potentiometric data. The values of β_{112} at different ionic strengths together with the values of literature are shown in Table 5.

The dependence of the dissociation and stability constants on the ionic strength can be described according to the previous works[14−17,25]

$$\log \beta_{112}(I) = \log \beta_{112}(I_1) - AZ^*\left(\frac{I^{0.5}}{1 + BI^{0.5}} - \frac{I_1^{\ 0.5}}{1 + BI_1^{\ 0.5}}\right) + C(I - I_1) + D(I^{1.5} - I_1^{\ 1.5}) \tag{12}$$

where *I* and I_1 are the actual and reference ionic strengths, respectively, and according to eq 13

$$p\mathrm{M}^{m+} + q\mathrm{L}^{n-} + r\mathrm{H}^+ \rightleftarrows (\mathrm{M}_p\mathrm{L}_q\mathrm{H}_r)^{pm-qn+r} \tag{13}$$

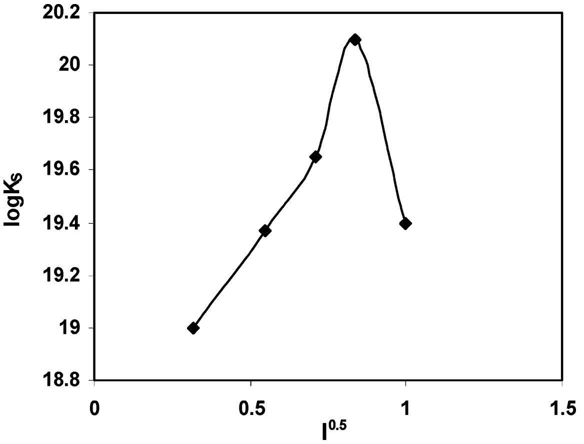

Figure 2. Plot of log K_S for WO_3L^{3-} vs the square root of ionic strength.

Table 6. Parameters for the Dependence on the Ionic Strength of Dissociation and Stability Constants at 25 °C

species	C	D	Z^*
K_3	1.057	−0.864	6
K_2	0.803	−0.730	4
K_1	0.210	−0.775	2
WO_3L^{3-}	7.130	−5.465	6

Table 7. Calculated Stability Constants at pH 7.5 and Different Ionic Strengths for the Complexation of Tungsten(VI) with NTA, T = 25 °C

I/mol dm^{-3}	log β_{112}
0.1	18.94
0.3	19.38
0.5	19.65
0.7	19.72
1.0	19.51

$Z^* = pm^2 + qn^2 + r - (pm + qn + r)^2$, where m and n are the charges on the metal ion and the ligand, respectively. Considering $A = 0.5115$ and $B = 1.489$, eq 12 can be simplified

$$\log \beta_{112}\,(I) = \log \beta_{112}\,(I_1) - Z^*\left(\frac{I^{0.5}}{2 + 3I^{0.5}} - \frac{I_1^{\,0.5}}{2 + 3I_1^{\,0.5}}\right) + C(I - I_1) + D(I^{1.5} - I_1^{\,1.5}) \quad (14)$$

where C and D are empirical coefficients and their values were obtained by minimizing the error squares sum, (U), and the Gauss−Newton nonlinear least-squares method in a suitable computer program

$$U = \sum_i (a_i - b_i)^2 \qquad (i = 1, 2, 3, ...) \qquad (15)$$

where a is a quasiexperimental quantity and b_i is a calculated one. The values of C and D are shown in Table 6. In this research, a_i is the experimental stability constant and b_i is the calculated one. We have used $I_1 = 0.5$ as the reference ionic strength in order to obtain better consistency between experimental and calculated stability constants. The calculated stability constants are given in Table 7.

The dependence on ionic strength of log β_{112} is shown in Table 4, which is in good agreement with the results obtained in the previous papers.[14−17,25] The dependence of β_{112} on the ionic strength is shown in Figure 2. This figure shows that β_{112} has one maximum at $I = 0.7$ similar to the maximum for the Mo(VI)−NTA system in our previous paper.[25]

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