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Ionic Strength Dependence of Dioxovanadium(V) Complexation with Ethylene Glycol-bis(2-aminoethylether)-*N*,*N*,*N*′,*N*′-tetraacetic Acid

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ABSTRACT: Stability constants for the complexation processes of the VO_2^+ ion with ethylene glycol-bis(2-aminoethylether)-*N*,*N*,*N*′,*N*′-tetraacetic acid (EGTA) in sodium perchlorate aqueous solutions for ionic strengths between (0.10 and 1.00) $mol \cdot dm^{-3}$ were calculated at $T = 298$ K by potentiometric and UV spectroscopic methods. The results indicate that EGTA forms the VO_2H_3L, $VO_2H_2L^-$, and VO_2HL^{2-} species with the VO_2^+ ion in the pH range of about 1.00 to 2.50. Different thermodynamic models (extended Debye−Hückel type (EDH), specific ion interaction theory (SIT), and parabolic) have been examined for the ionic strength dependence of the stability and dissociation constants for the above-mentioned species.

INTRODUCTION

Several reports have been published regarding the existence of vanadium in biological systems.[1] Vanadium compounds may act as potential antimetastatic agents by inhibiting the induction of intracellular adhesive molecules[2] and may also induce cytotoxic effects through DNA cleavage/fragmentation and plasma membrane lipoperoxidation.[1]

Ethylene glycol-bis(2-aminoethylether)-*N*,*N*,*N*′,*N*′-tetraacetic acid (EGTA) can induce the rapid and extensive release of calcium from energized molecules[3] and activates Ca^{2+}-stimulated ATPase of rat liver plasma membranes.[4] It is used as a buffer component in dideoxy sequencing of RNA using reverse transcriptase.[5]

Following our previous works on the complexation of dioxovanadium(V), molybdenum(VI), and tungsten(VI) complexes with complexones at different ionic strengths by using the extended Debye−Hückel type (EDH), specific ion interaction theory (SIT), and parabolic models and in different aqueous solutions of methanol,[6−14] the influence of ionic strength on the VO_2^+−EGTA stability constants have been studied for ionic strengths between (0.10 and 1.00) $mol \cdot dm^{-3}$ in this paper. Finally the results have been compared with literature data, though few data were available.

EXPERIMENTAL SECTION

Reagents. All chemicals were of analytical reagent grade. Perchloric acid, sodium hydroxide, potassium hydrogen carbonate, sodium perchlorate, hydrochloric acid, sodium carbonate, and sodium monovanadate were purchased from Merck and EGTA from Fluka and were used without further purification. The NaOH solutions were prepared from titrisol solutions, and their concentrations were determined by several titrations with standard HCl. The HCl solution was standardized with sodium carbonate solution (Na_2CO_3).[6] Dilute perchloric acid solution was standardized against $KHCO_3$.[15] A stock solution of vanadium(V) was prepared by dissolution of anhydrous sodium monovanadate in perchloric acid solution to prevent the formation of the decavanadate. The solution stood overnight before use to obtain only the VO_2^+ ion, and isopolyvanadates will not be formed or if small amounts still exist they will be decomposed. In all experiments double-distilled water with a specific conductance equal to (1.3 ± 0.1) $\mu S \cdot cm^{-1}$ was used.

Measurements. All measurements were carried out at $T = 298$ K. A Metrohm pH meter, 827, was used for pH measurements. The hydrogen ion concentration was measured with a Metrohm combination electrode, model 6.0228.010. A 0.01 $mol \cdot dm^{-3}$ perchloric acid solution containing 0.09 $mol \cdot dm^{-3}$ sodium perchlorate (for adjusting the ionic strength to 0.10 $mol \cdot dm^{-3}$) was used as a standard solution of hydrogen ion concentration. The same procedure was repeated for the other ionic strengths.[15] The calibration has been done for the whole pH ($pH = -\log[H^+]$) range used. The change in liquid junction potential[15] was calculated from eq 1

$$pH(real) = pH(measured) + a + b[H^+](measured) \qquad (1)$$

a and b were determined by measurement of the hydrogen ion concentration for two different solutions of $HClO_4$ with sufficient $NaClO_4$ to adjust the ionic media.[15] Spectrophotometric measurements were performed with a Varian Cary 300 UV−vis spectrophotometer with a Pentium 4 computer between (245 and 280) nm in thermoregulated matched 10 mm quartz cells. The measurement cell was of the flow type. Circulation of the complex solution from the potentiometric cell to the spectrophotometric cell was possible by using a Masterflux pump. Therefore the pH and absorbance of the solution could be measured simultaneously.

Different metal−ligand concentrations and ligand−metal molar ratios were tested. Finally a good fit and the speciation pattern and minimum error function have been obtained with $C_L = 1.29 \cdot 10^{-3}$ and $C_{VO_2} = 6.00 \cdot 10^{-4}$ $mol \cdot dm^{-3}$. The 50 cm^3 acidic solutions of dioxovanadium(V) ($6.00 \cdot 10^{-4}$ $mol \cdot dm^{-3}$) were titrated with basic solutions of EGTA ($1.29 \cdot 10^{-3}$ $mol \cdot dm^{-3}$) at a fixed ionic strength. The absorbance of the solution was measured after each addition and adjusting the pH. The titration and absorbance measurement were repeated each time for the other ionic strengths up to $I = 1.00$ $mol \cdot dm^{-3}$ of sodium perchlorate. According to the literature,[16] in acidic solution

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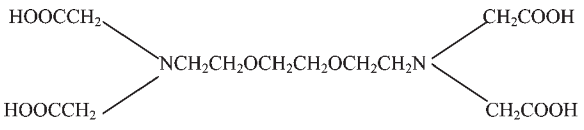
(pH < 2.5) and in the presence of a large excess of ligand, vanadium(V) exists as the VO_2^+ ion. In all cases, the measurement was repeated at least three times, and the resulting average values and corresponding standard deviations are shown in the text and tables.

■ RESULTS AND DISCUSSION

Complexation of Dioxovanadium(V) with EGTA. *Dissociation Constants.* The chemical structure of EGTA is shown below:



The equilibria (2) are studied where L^{n-} represents the fully deprotonated ligand.

$$H_iL^{(n-i)-} \rightleftharpoons H^+ + H_{i-1}L^{(n+1-i)-}$$

$$K_i = \frac{[H^+][H_{i-1}L^{(n+1-i)-}]}{[H_iL^{(n-i)-}]} \qquad (2)$$

Four values of dissociation constants of EGTA which were obtained by using the potentiometric technique and the Microsoft Excel 2000 program are gathered in Table 1 together with literature values. Three titrations have been done for each ionic strength, and approximately 200 points have been used in the calculations at each ionic strength.[17]

Speciation Studies. Different stoichiometric models were tested. Assuming three species, VO_2H_3L, $VO_2H_2L^-$, and VO_2HL^{2-}, was our accepted model on the basis of suitable speciation and fitting diagrams. Stability constants were calculated from the summation of dissociation and formation constant values. For the formation of dioxovanadium(V)−EGTA complexes the stability constant of $(VO_2)_pH_qL_r^{(p+q-4r)}$, β_{pqr}, is defined as follows

$$pVO_2^+ + qH^+ + rL^{4-} \rightleftharpoons (VO_2)_pH_qL_r^{(p+q-4r)}$$

$$\beta_{pqr} = \frac{[(VO_2)_pH_qL_r^{(p+q-4r)}]}{[VO_2^+]^p[H^+]^q[L^{4-}]^r} \qquad (3)$$

The simultaneous existence of three species is similar to the complexation of UO_2^{2+} with ethylenediaminetetraacetic acid (EDTA) which has been reported in the literature.[18] Three species, $UO_2H_4L^{2+}$, $UO_2H_3L^+$, and UO_2H_2L, were assumed on the basis of satisfactory graphical and numerical fitting.[18]

The absorbance data in the UV range (255 to 280) nm were used for minimizing the error function on the basis of a Gauss−Newton nonlinear least-squares method in the Microsoft Excel 2000 program according to the function $A = f(\text{pH})$. The error function is defined as[6,8]

$$U = \sum (A_{exp} - A_{cal})^2 \qquad (4)$$

A_{exp} values have been gathered from the UV spectrophotometric measurements and are reported at different pH and wavelengths in Tables A1 to A5. A_{cal} values have been obtained for the main model which was discussed above. The concentration of the complexes contributing to the general species distribution in our experimental conditions were calculated by the Microsoft Excel 2000 program. Complexes with negligible concentrations over the entire range of experimental conditions are not taken into account in our proposed results. Finally the best fit and minimum error function were obtained with the VO_2H_3L, $VO_2H_2L^-$, and VO_2HL^{2-} species. A_{exp} and A_{cal} values at T = 298 K, I = 0.1 mol·kg^{-1}, and 270 nm are shown in Figure 1 which shows a very good graphical fit. Similar fits have been obtained for the other ionic strengths. The chosen model is also in close agreement with experimental data. The speciation diagrams are shown in Figure 2 for different ionic strengths. A_{cal} values have been determined from the combination of the following mass-balance and Beer−Lambert laws for our accepted model (L = EGTA)

$$A = \varepsilon_0[VO_2^+] + \varepsilon_{VO_2H_3L}[VO_2H_3L] + \varepsilon_{VO_2H_2L^-}[VO_2H_2L^-] + \varepsilon_{VO_2HL^{2-}}[VO_2HL^{2-}] \qquad (5)$$

$$C_{VO_2^+} = [VO_2^+] + [VO_2H_3L] + [VO_2H_2L^-] + [VO_2HL^{2-}] \qquad (6)$$

$$C_L = [VO_2H_3L] + [VO_2H_2L^-] + [VO_2HL^{2-}] + [H_4L] + [H_3L^-] + [H_2L^{2-}] + [HL^{3-}] \qquad (7)$$

Table 1. Dissociation Constants K_4, K_3, K_2, and K_1 of EGTA at Different Ionic Strengths, I, of $NaClO_4$ and T = 298 K

I / mol·dm^{-3}	I / mol·kg^{-1}	log K_4(M)	log K_4(m)	log K_3(M)	log K_3(m)	log K_2(M)	log K_2(m)	log K_1(M)	log K_1(m)	ref
0.10	0.10	9.72 ± 0.05	9.73	8.73 ± 0.09	8.74	2.76 ± 0.02	2.77	2.05 ± 0.07	2.06	this work
0.30	0.30	9.52 ± 0.02	9.54	8.54 ± 0.12	8.56	2.72 ± 0.06	2.74	1.92 ± 0.05	1.94	this work
0.50	0.51	9.37 ± 0.03	9.40	8.38 ± 0.08	8.41	2.66 ± 0.02	2.69	1.83 ± 0.12	1.86	this work
0.70	0.72	9.55 ± 0.06	9.60	8.60 ± 0.02	8.65	2.75 ± 0.02	2.80	2.00 ± 0.03	2.05	this work
1.00	1.05	9.77 ± 0.08	9.84	8.78 ± 0.01	8.85	2.81 ± 0.05	2.88	2.11 ± 0.06	2.18	this work
0.1		9.51		8.92		2.82		2.46		38[a]
0.1		9.54		8.93		2.73		2.08		39[b]
0.1		9.53		8.88						40[c]
0.1		9.46		8.85		2.65		2.0		41[d]
0.1		9.60		8.79		2.67		2.04		19[e]
0.5		9.22		8.39		2.49		2.01		19[f]
0.5		8.89		8.40		2.50		0.93		42[g]

[a] KNO_3. [b] KNO_3. [c] KCl. [d] KNO_3, T = 293 K. [e] NaCl. [f] NaCl. [g] $NaClO_4$.

and the formation constants

$$VO_2{}^+ + H_4L \rightleftharpoons VO_2H_3L + H^+ K_{VO_2H_3L} = \frac{[VO_2H_3L][H^+]}{[VO_2{}^+][H_4L]} \tag{8}$$

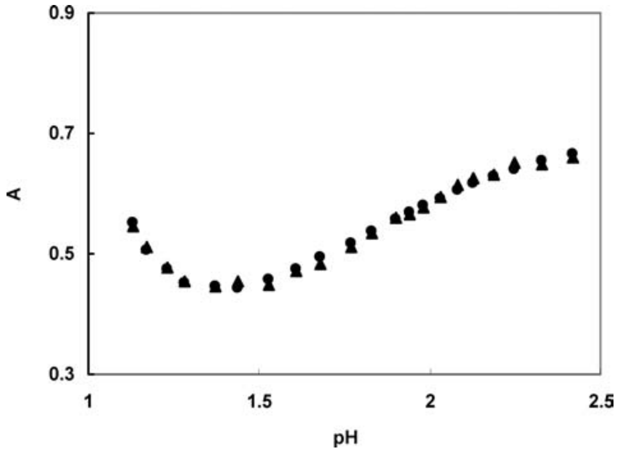


Figure 1. A_{exp} and A_{cal} values at $T = 298$ K, $I = 0.10$ mol·kg^{-1}, and 270 nm. ●, A_{cal}; ▲, A_{exp} for the model including VO_2H_3L, $VO_2H_2L^-$, and VO_2HL^{2-}.

$$VO_2H_3L \rightleftharpoons VO_2H_2L^- + H^+ K_{VO_2H_2L^-} = \frac{[VO_2H_2L^-][H^+]}{[VO_2H_3L]} \tag{9}$$

$$VO_2H_2L^- \rightleftharpoons VO_2HL^{2-} + H^+ K_{VO_2HL^{2-}} = \frac{[VO_2HL^{2-}][H^+]}{[VO_2H_2L^-]} \tag{10}$$

where $C_{VO_2{}^+}$ and C_L are the total concentration of $VO_2{}^+$ and the ligand, respectively.

The average values of the experimental and calculated stability constants at various wavelengths are gathered in Tables 2 and 3. It is important to note that, according to the values of the dissociation constants of EGTA (Table 1), it is reasonable to assume protonated species for this complexation reaction.

Comparison with Literature Data. The authors are not aware of any previous publication of the stability constants for the complexation of $VO_2{}^+$ ion with EGTA in different sodium perchlorate aqueous solutions. The most recently published data are about the formation constants for the complexation of Ca^{2+} and Mg^{2+} with EGTA at different ionic strengths; log β_{111} = 3.56 (Mg^{2+}-EGTA) and log β_{111} = 5.27 (Ca^{2+}-EGTA) at $I = 0.1$ mol·dm^{-3} of KCl and $T = 298$ K.[19] We have recently published stability constants for this complexation reaction in different water + methanol mixtures which showed that only two species, VO_2H_3L

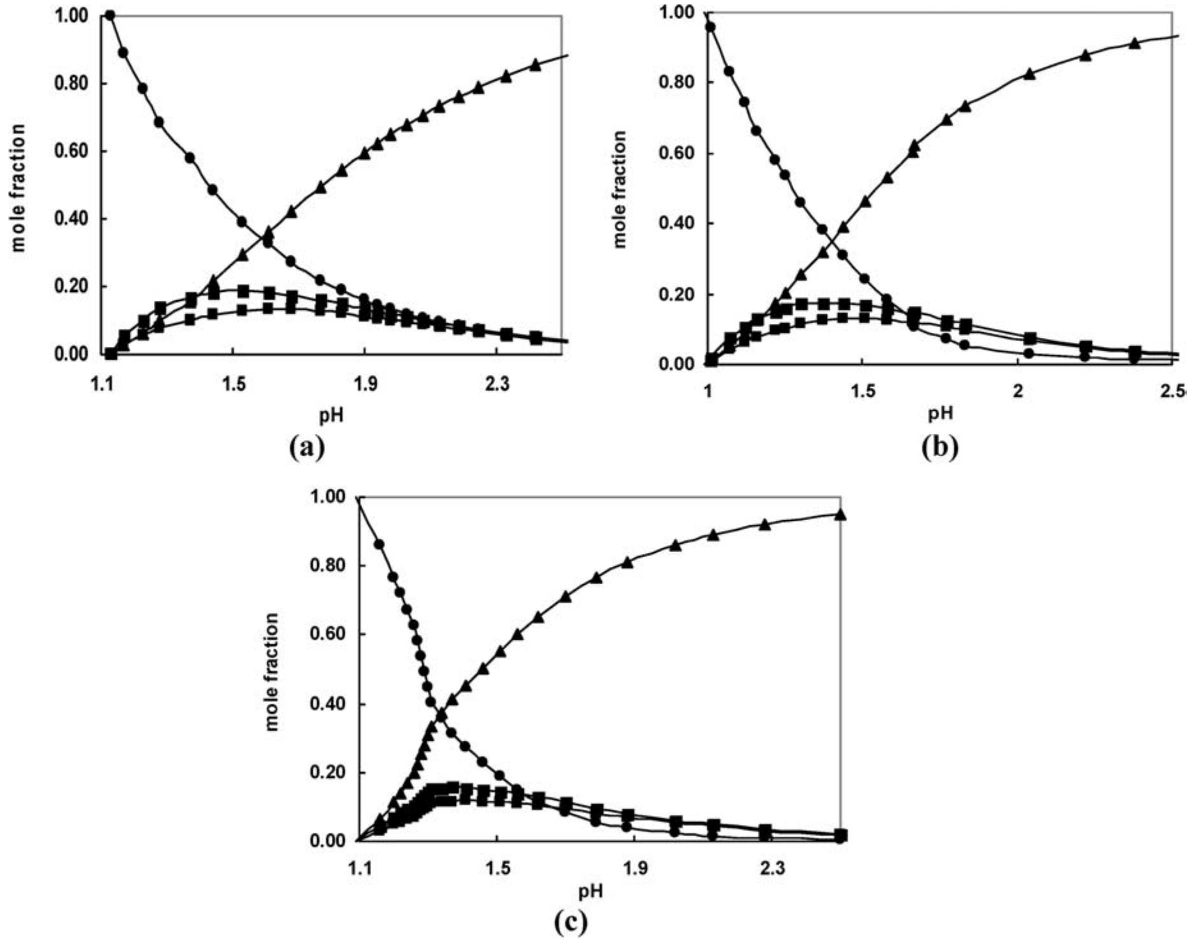


Figure 2. Speciation diagrams at $T = 298$ K (a) $I = 0.10$ mol·kg^{-1}, (b) $I = 0.51$ mol·kg^{-1}, and (c) $I = 1.05$ mol·kg^{-1} for the model including VO_2H_3L, $VO_2H_2L^-$, and VO_2HL^{2-}. $C_{VO2}{}^+ = 6.00 \cdot 10^{-4}$ mol·dm^{-3} and $C_L = 1.29 \cdot 10^{-3}$ mol·dm^{-3}. ●, $VO_2{}^+$; ▲, VO_2HL^{2-}; ■, VO_2H_3L; small ■, $VO_2H_2L^-$.

and $VO_2H_2L^-$, exist in the solution with stability constants in the range of about (25.75 ± 0.12) to (29.99 ± 0.10).[20]

Ionic Strength Dependence of Dissociation and Stability Constants According to the SIT, Parabolic, and EDH Models. Interactions between the different components in a given system are a direct function of their activity coefficients, and these depend on the ionic medium and ionic strength.[21] Most experimental studies of chemical equilibria are performed according to the constant ionic medium method which means in the presence of an excess of an inert electrolyte (in this research sodium perchlorate). In this way, the activity coefficients of all species are constant over a broad concentration range.[21] The activity coefficient γ_j of an ion j of charge z_j in a solution of ionic strength I may be described by[22,23]

$$\log \gamma_j = \frac{-z_j^2 0.509\sqrt{I}}{1 + 1.5\sqrt{I}} + \sum_k \varepsilon(j, k, I) m_k \qquad (11)$$

The summation in eq 11 extends over all ions k present in solution at the molality m_k. Activity coefficients for the neutral species are given by the linear relationship:

$$\log \gamma = k_m I \qquad (12)$$

where k_m is the salting out coefficient (Setschenow coefficient[24]) of a neutral species. For correct application of the SIT, concentrations should be reported in the molal concentration scale. Molar ionic strengths have been converted to the molal scale according to the literature.[25] log β values were converted to the molal concentration scale according to the following equation[6]

$$\log \beta(m) = \log \beta(M) + \sum \nu \log(m/M) \qquad (13)$$

where m and M stand for molality and molarity, respectively. $\Sigma\nu$ is the sum of stoichiometric coefficients of the reaction species. Equation 11, which is the basis of the SIT, is a simplified version of the Pitzer ion-interaction approach, neglecting triple interactions which are important only in very concentrated solutions and the interactions between ions of the same sign. The first term in eq 11 shows the nonspecific long-range contribution of the electrostatic region, the second reflects the contribution due to short-range ion−ion, ion−neutral, or neutral−neutral interactions, in accordance with the Bronsted postulate that $\varepsilon(j, k) = 0$ if ions have the same charge sign. The rationale behind this is that ε, which describes specific short-range interactions, must be small for ions of the same charge since they are usually far from one another due to electrostatic repulsion. This holds to a lesser extent also for uncharged species.

There are many papers in the literature about the SIT.[26−32] Sammartano et al.[32] proposed a modified version of the SIT approach, in which the specific coefficients are expressed as a function of I

$$\varepsilon = \varepsilon_\infty + (\varepsilon_0 - \varepsilon_\infty)(I + 1)^{-1} \qquad (14)$$

where ε_∞ is the value of ε for $I \rightarrow \infty$ and ε_0 is the value of ε for $I \rightarrow 0$. ε_∞ could be defined as follows

$$\varepsilon_\infty = \varepsilon_\infty{}^{(0)} + \varepsilon_\infty{}^{(1)} I \qquad (15)$$

The results were satisfactory in the whole range $0.1 < I/\text{mol}\cdot\text{kg}^{-1} < 6$.[21] Equation 14 comes from the need for adding a term which takes into account the formation of weak (or even very weak) ion pairs.[21]

Ciavatta[23] proposed another expression for the ionic strength dependence of ε

$$\varepsilon = \varepsilon^{(0)} + \varepsilon^{(1)} \ln(1 + I) \qquad (16)$$

Under our conditions and according to previous work,[6,8] it is often possible to simplify the summation in eq 11 so that only ion interaction coefficients between the participating ionic species and the ionic medium ions are included as shown in eqs 17 to 20. The stability constant of $(VO_2)_pH_qL_r^{(p+q-4r)}$, β_{pqr}, determined in an ionic medium (1:1 salt $NaClO_4$) of ionic strength I, is related to the corresponding value at zero ionic strength, $\beta^0{}_{pqr}$, by eq 17

$$\log \beta_{pqr} - \Delta z^2 \text{DH} = \log \beta^0{}_{pqr} - \Delta\varepsilon I \qquad (17)$$

where

$$\Delta z^2 = (p + q - 4r)^2 - (p + q + 16r) \qquad (18)$$

$$\text{DH} = \frac{0.509\sqrt{I}}{1 + 1.5\sqrt{I}} \qquad (19)$$

$$\Delta\varepsilon = \varepsilon((VO_2)_pH_qL_r{}^{(p+q-4r)}, \text{Na}^+ \text{ or } ClO_4{}^-) - \varepsilon(VO_2{}^+, ClO_4{}^-) - q\varepsilon(H^+, ClO_4{}^-) - \varepsilon(L^{4-}, Na^+) \qquad (20)$$

Equilibria involving $H_2O(l)$ as a reactant or product require a correction for the activity of water.[22] In most experimental studies of equilibria in dilute aqueous solutions, where an ionic medium is

Table 2. Average Experimental and Calculated Values of log β_{131} at pH = 1.00 to 2.50 and Different Ionic Strengths for the Complexation of Dioxovanadium(V) with EGTA, T = 298 K

I	I	log β_{131}(M)		log β_{131}(m)	
$\text{mol}\cdot\text{dm}^{-3}$	$\text{mol}\cdot\text{kg}^{-1}$	exptl	calcd	exptl	calcd
0.10	0.10	25.90 ± 0.09	25.90 ± 0.00	25.93	25.93 ± 0.00
0.30	0.30	25.12 ± 0.05	25.30 ± 0.94	25.16	25.36 ± 0.93
0.50	0.51	25.33 ± 0.20	25.43 ± 2.04	25.40	25.50 ± 2.05
0.70	0.72	26.22 ± 0.40	25.94 ± 3.27	26.31	26.02 ± 3.31
1.00	1.05	27.08 ± 0.10	27.18 ± 5.29	27.21	27.31 ± 5.46

Table 3. Average Experimental and Calculated Values of log β_{121} and log β_{111} at pH = 1.00 to 2.50 and Different Ionic Strengths for the Complexation of Dioxovanadium(V) with EGTA, T = 298 K

I	I	log β_{121}(M)		log β_{121}(m)		log β_{111}(M)		log β_{111}(m)	
$\text{mol}\cdot\text{dm}^{-3}$	$\text{mol}\cdot\text{kg}^{-1}$	exptl	calcd	exptl	calcd	exptl	calcd	exptl	calcd
0.10	0.10	24.70 ± 0.06	24.70 ± 0.00	24.72	24.72 ± 0.00	23.49 ± 0.20	23.49 ± 0.00	23.51	23.51 ± 0.00
0.30	0.30	24.01 ± 0.25	24.22 ± 1.05	24.05	24.27 ± 1.03	22.76 ± 0.10	23.11 ± 1.36	22.79	23.15 ± 1.34
0.50	0.51	24.34 ± 0.35	24.44 ± 2.27	24.40	24.50 ± 2.25	23.36 ± 0.15	23.40 ± 2.96	23.40	23.45 ± 2.93
0.70	0.72	25.33 ± 0.30	25.02 ± 3.63	25.41	25.10 ± 3.64	24.43 ± 0.35	24.07 ± 4.73	24.49	24.12 ± 4.74
1.00	1.05	26.26 ± 0.20	26.37 ± 5.87	26.37	26.48 ± 6.01	25.45 ± 0.45	25.60 ± 7.66	25.54	25.68 ± 7.82

used in large excess with respect to the reactants, the activity of water is near constant and equal to 1.[22] According to eq 18, $\Delta z^2 = -20, -18, -14$ for the VO_2H_3L, $VO_2H_2L^-$, and VO_2HL^{2-}, complexes respectively.[22] The linear regressions were done on the basis of eq 17, the values of $\Delta\varepsilon = (-3.60 \pm 1.44, -3.82 \pm 1.50$, and $-3.91 \pm 1.84)$ $kg \cdot mol^{-1}$ have been obtained for VO_2H_3L, $VO_2H_2L^-$, and VO_2HL^{2-}, respectively. $\varepsilon(H^+, ClO_4^-) = (0.14 \pm 0.02)$ $kg \cdot mol^{-1}$,[22] thus, the following equations are valid[6]

$$\varepsilon(VO_2^+, ClO_4^-) + \varepsilon(L^{4-}, Na^+) = 3.18 \pm 1.44 \qquad (21)$$

$$\varepsilon(VO_2H_2L^-, Na^+) = -0.36 \pm 2.08 \qquad (22)$$

$$\varepsilon(VO_2HL^{2-}, Na^+) = -0.59 \pm 2.33 \qquad (23)$$

The values of $\log \beta^0{}_{131}$, $\log \beta^0{}_{121}$, and $\log \beta^0{}_{111}$ have been obtained: 27.40 ± 0.91, 25.93 ± 0.95, and 24.18 ± 1.16. In this paper, the classic SIT model (eq 11) was used instead of the two parameter SIT model proposed in a previous paper by Sammartano,[21] owing to the short ionic strength interval investigated here.[30]

The parabolic model has also been applied to the dependence of metal complex formation and dissociation constants on ionic strength[29]

$$\log \beta_{pqr} - \Delta z^2 DH = \log \beta^0{}_{pqr} - \Delta\varepsilon I - \Delta\delta i I^2 \qquad (24)$$

The parabolic model with two coefficients is satisfactory for ionic strengths to $I = 14$ m.[33] The parabolic model because of its advantages in mathematical simplicity, and its less-parametrized nature is comparable to the Pitzer model in many cases.

Deviations from the Debye−Hückel equation could be due to a term linear in ionic strength, ion association, or both phenomena taken together. The dependence of the dissociation and stability constants on ionic strength according to the extended Debye−Hückel-type equation (EDH) can be described as follows[34−36]

$$\log \beta(I) = \log \beta(I_1) - Z^* \left(\frac{I^{0.5}}{1.955 + 2.91 I^{0.5}} - \frac{I_1{}^{0.5}}{1.955 + 2.91 I_1{}^{0.5}} \right) + C(I - I_1) + D(I^{1.5} - I_1{}^{1.5}) \qquad (25)$$

where I and I_1 are the actual and reference ionic strengths, respectively and according to reaction 3, $Z^* = p + 16r + q - (p + q - 4r)^2$. For $I < 2$ $mol \cdot dm^{-3}$ it was not necessary to include the EI^2 quadratic term in eq 25. C and D are empirical coefficients, and their values were obtained by minimizing the error squares sum, (U), using the Gauss−Newton nonlinear least-squares method in the Excel 2000 program:

$$U = \sum_i (a_i - b_i)^2 \qquad (i = 1, 2, 3, ...) \qquad (26)$$

where a_i is a quasi-experimental quantity and b_i is a calculated one. It has been reported that for some carboxylic acids (malic, tartaric, 1,2,3-propanetricarboxylic, and 1,2,3,4-butanetetracarboxylic acids) in different background salts (NaCl, KCl, and Me_4NCl), the C and D parameters proved independent of the acid and dependent only on anion charge.[37] Sammartano defined C as an empirical parameter that can be expressed as:[31]

$$C = c_\infty + (c_0 - c_\infty)(I + 1)^{-1} \qquad (27)$$

where c_∞ and c_0 are the values of C at $I \rightarrow \infty$ and $I \rightarrow 0$, respectively. The values of C and D are shown in Table 4. The values of C and D have been inserted in eq 25, then the values of the calculated stability constants have been determined according to EDH, and their values are gathered in Tables 2 and 3. We have used $I_1 = 0.1$ as the reference ionic strength to obtain better consistency between experimental and calculated stability constants.

Values of $\log \beta^0$, $\log K^0$, $\Delta\varepsilon$, $\Delta\delta$, and R^2 on the basis of EDH, SIT, and parabolic models for the three species, VO_2H_3L, $VO_2H_2L^-$, and VO_2HL^{2-}, and the dissociation constants are summarized in Tables 5 and 6. Parabolic model gives best fits.

Table 4. Parameters for the Dependence on the Ionic Strength of Dissociation and Stability Constants at T = 298 K

	C	D		
species	$kg \cdot mol^{-1}$	$kg^{1.5} \cdot mol^{-1.5}$	Z^*	ref
K_1	-1.05 ± 0.66	1.27 ± 0.65	2	this work
K_2	0.59 ± 0.40	-0.07 ± 0.39	4	this work
K_3	-0.68 ± 0.86	1.27 ± 0.84	6	this work
K_4	-0.16 ± 0.70	0.97 ± 0.69	8	this work
VO_2H_3L	-1.63 ± 2.76	4.61 ± 2.71	20	this work
$VO_2H_2L^-$	-1.44 ± 3.04	4.62 ± 2.98	18	this work
VO_2HL^{2-}	-2.54 ± 3.96	5.64 ± 3.87	14	this work

Table 5. Values of $\log \beta^0$, $\Delta\varepsilon$, $\Delta\delta$, and R^2 According to Debye−Hückel, SIT, and Parabolic Models

			$\Delta\varepsilon$	$\Delta\delta$	
species	model	$\log \beta^0$(m)	$kg \cdot mol^{-1}$	$kg^2 \cdot mol^{-2}$	R^2
VO_2H_3L	Debye−Hückel	26.01 ± 0.21			0.94
$VO_2H_2L^-$	Debye−Hückel	24.73 ± 0.24			0.95
VO_2HL^{2-}	Debye−Hückel	23.41 ± 0.31			0.93
VO_2H_3L	SIT	27.40 ± 0.91	-3.60 ± 1.44		0.96
$VO_2H_2L^-$	SIT	25.93 ± 0.95	-3.82 ± 1.50		0.96
VO_2HL^{2-}	SIT	24.18 ± 1.16	-3.91 ± 1.84		0.94
VO_2H_3L	Parabolic	27.84 ± 0.13	-1.34 ± 0.14	-1.95 ± 0.16	0.98
$VO_2H_2L^-$	Parabolic	26.37 ± 0.10	-1.57 ± 0.12	-1.94 ± 0.15	0.98
VO_2HL^{2-}	Parabolic	24.68 ± 0.17	-1.30 ± 0.21	-2.25 ± 0.22	0.96

Table 6. Values of $\log K^0$, $\Delta\varepsilon$, $\Delta\delta$, and R^2 According to Debye−Hückel, SIT, and Parabolic Models

			$\Delta\varepsilon$	$\Delta\delta$	
species	model	$\log K^0$(m)	$kg \cdot mol^{-1}$	$kg^2 \cdot mol^{-2}$	R^2
K_1	Debye−Hückel	2.12 ± 0.05			0.86
K_2	Debye−Hückel	2.80 ± 0.03			0.86
K_3	Debye−Hückel	8.85 ± 0.07			0.88
K_4	Debye−Hückel	9.85 ± 0.06			0.92
K_1	SIT	2.16 ± 0.25	-0.36 ± 0.40		0.73
K_2	SIT	3.17 ± 0.08	-0.51 ± 0.13		0.98
K_3	SIT	9.25 ± 0.29	-0.74 ± 0.46		0.90
K_4	SIT	10.46 ± 0.23	-0.91 ± 0.37		0.95
K_1	Parabolic	3.16 ± 0.01	-0.55 ± 0.02	0.04 ± 0.04	0.98
K_2	Parabolic	2.30 ± 0.07	0.35 ± 0.10	-0.61 ± 0.13	0.92
K_3	Parabolic	9.40 ± 0.01	0.03 ± 0.02	-0.66 ± 0.06	0.96
K_4	Parabolic	10.58 ± 0.02	-0.30 ± 0.04	-0.53 ± 0.05	0.98

CONCLUSIONS

Several factors can affect the stability constants of the VO_2^+ + EGTA system, such as: (a) medium stabilization which means the interaction of negatively charged complex species with the cation of the supporting electrolyte, (b) the charge on the complex, and (c) etheric oxygens.[30] Sammartano et al.[30] confirmed the stabilization of metal−ligand species by the presence of etheric oxygens in the ligand structure for the complexation of dioxouranium(VI) with some dicarboxylic ligands in NaCl and KNO_3 solutions. It seems that the etheric oxygens are also very important in this work for the stability constants of the EGTA complexes at different ionic strengths, and therefore we have large stability constant values.

Figure 2a shows that VO_2H_3L reaches a maximum of 19 % at $I = 0.10\ \mathrm{mol \cdot kg^{-1}}$, 270 nm, and pH = 1.53, but this occurs at pH < 1.53 for the other ionic strengths. A similar trend exists for the $VO_2H_2L^-$ species. Regarding the dissociation constant values at infinite dilution, there was more or less a good agreement between the three models. Approximately there is a good agreement between the log β^0 values obtained from the SIT and parabolic models, but the results are different from the Debye−Hückel. There was a large difference between $\Delta\varepsilon$ values which were obtained in the SIT and parabolic models. Large uncertainties exist for the dissociation and stability constant values (except K_1) on the basis of C values. The data fit with the Debye−Hückel (on the basis of the errors for C and D) and errors for the SIT, and parabolic models show that parabolic model is the best.

APPENDIX

A_{exp} values have been gathered from the UV spectrophotometric measurements and are reported at different pH and wavelengths in Tables A1 to A5.

Table A1. Experimental Values of Absorbance at Different pH and Wavelengths at T = 298 K, I = 0.10 mol·kg^{-1}

	λ/nm					
pH	255	260	265	270	275	280
1.13	0.5927	0.5657	0.5606	0.5460	0.5238	0.5127
1.17	0.5548	0.5356	0.5233	0.5109	0.4825	0.4713
1.23	0.5299	0.5041	0.5003	0.4763	0.4626	0.4418
1.28	0.5091	0.4832	0.4701	0.4532	0.4350	0.4226
1.37	0.4966	0.4692	0.4621	0.4447	0.4230	0.4034
1.44	0.5178	0.4925	0.4650	0.4539	0.4179	0.3993
1.53	0.5307	0.5034	0.4706	0.4485	0.4254	0.3953
1.61	0.5677	0.5273	0.4894	0.4718	0.4344	0.4041
1.68	0.6013	0.5493	0.5184	0.4842	0.4433	0.4216
1.77	0.6588	0.6057	0.5508	0.5127	0.4757	0.4370
1.83	0.6912	0.6315	0.5742	0.5348	0.4852	0.4503
1.90	0.7293	0.6580	0.6063	0.5591	0.5091	0.4657
1.94	0.7470	0.6852	0.6180	0.5663	0.5170	0.4692
1.98	0.7729	0.7005	0.6283	0.5761	0.5254	0.4779
2.03	0.7981	0.7213	0.6522	0.5956	0.5395	0.4873
2.08	0.8290	0.7427	0.6724	0.6137	0.5513	0.5008
2.13	0.8539	0.7568	0.6875	0.6268	0.5569	0.5085
2.19	0.8599	0.7683	0.6920	0.6326	0.5705	0.5167
2.25	0.8876	0.7895	0.7149	0.6520	0.5818	0.5219
2.33	0.8973	0.8000	0.7270	0.6493	0.5834	0.5341
2.42	0.9120	0.8227	0.7313	0.6609	0.5911	0.5304
2.54	0.9335	0.8331	0.7470	0.6686	0.6065	0.5434

Table A2. Experimental Values of Absorbance at Different pH and Wavelengths at T = 298 K, I = 0.30 mol·kg^{-1}

	λ/nm					
pH	255	260	265	270	275	280
1.24	0.5916	0.5733	0.5536	0.5624	0.5240	0.5085
1.30	0.5730	0.5460	0.5358	0.5296	0.5052	0.4757
1.36	0.5428	0.5279	0.5052	0.5008	0.4725	0.4509
1.43	0.5228	0.5087	0.4850	0.4822	0.4644	0.4273
1.52	0.5317	0.5162	0.4900	0.4743	0.4521	0.4213

Table A2. Continued

	λ/nm					
pH	255	260	265	270	275	280
1.61	0.5626	0.5311	0.4963	0.4924	0.4551	0.4191
1.68	0.5929	0.5560	0.5226	0.5007	0.4679	0.4328
1.76	0.6434	0.5965	0.5513	0.5290	0.4871	0.4492
1.86	0.6939	0.6426	0.5857	0.5579	0.5115	0.4645
1.96	0.7770	0.7052	0.6371	0.5999	0.5448	0.4896
2.02	0.8114	0.7395	0.6712	0.6175	0.5602	0.4997
2.09	0.8434	0.7672	0.6906	0.6410	0.5806	0.5185
2.17	0.8911	0.8018	0.7190	0.6697	0.6005	0.5367
2.22	0.9204	0.8237	0.7335	0.6729	0.6166	0.5427
2.29	0.9236	0.8408	0.7519	0.6855	0.6141	0.5510
2.36	0.9534	0.8715	0.7670	0.6993	0.6420	0.5640
2.44	0.9628	0.8762	0.7783	0.7044	0.6371	0.5876
2.53	0.9816	0.8893	0.7902	0.7207	0.6498	0.5770

Table A3. Experimental Values of Absorbance at Different pH and Wavelengths at T = 298 K, I = 0.51 mol·kg^{-1}

	λ/nm					
pH	255	260	265	270	275	280
0.99	0.5988	0.5778	0.5770	0.5583	0.5324	0.5196
1.01	0.5869	0.5653	0.5565	0.5398	0.5176	0.5083
1.07	0.5651	0.5374	0.5291	0.5193	0.4936	0.4783
1.12	0.5480	0.5240	0.5138	0.4972	0.4706	0.4664
1.16	0.5359	0.5061	0.5051	0.4895	0.4614	0.4483
1.22	0.5326	0.4996	0.4956	0.4707	0.4562	0.4480
1.25	0.5337	0.5088	0.4920	0.4794	0.4529	0.4428
1.30	0.5487	0.5174	0.5009	0.4771	0.4584	0.4379
1.37	0.5649	0.5313	0.5101	0.4883	0.4622	0.4333
1.44	0.5940	0.5590	0.5305	0.4953	0.4711	0.4439
1.51	0.6307	0.5911	0.5630	0.5128	0.4847	0.4649
1.58	0.6896	0.6284	0.5899	0.5495	0.5004	0.4773
1.66	0.7499	0.6856	0.6373	0.5861	0.5300	0.5010
1.67	0.8262	0.7484	0.6859	0.6254	0.5745	0.5273
1.77	0.8949	0.8102	0.7368	0.6651	0.6023	0.5576
1.83	0.9245	0.8338	0.7575	0.6772	0.6167	0.5660
2.04	0.9753	0.8664	0.7966	0.7148	0.6407	0.5816
2.22	0.9916	0.8940	0.8003	0.7190	0.6444	0.5897
2.38	0.9981	0.8907	0.8053	0.7226	0.6503	0.5872
2.60	1.0050	0.8956	0.8049	0.7166	0.6527	0.5897

Table A4. Experimental Values of Absorbance at Different pH and Wavelengths at T = 298 K, I = 0.72 mol·kg^{-1}

	λ/nm					
pH	255	260	265	270	275	280
1.05	0.5617	0.5514	0.5429	0.5159	0.4977	0.4780
1.11	0.5344	0.5176	0.5061	0.4834	0.4657	0.4352
1.14	0.5236	0.5117	0.4957	0.4635	0.4513	0.4321
1.16	0.5141	0.5065	0.4845	0.4704	0.4478	0.4204
1.17	0.5309	0.5163	0.5048	0.4750	0.4592	0.4457
1.18	0.5340	0.5173	0.4978	0.4655	0.4564	0.4330
1.19	0.5312	0.5043	0.5010	0.4611	0.4545	0.4310

Table A4. Continued

	λ/nm					
pH	255	260	265	270	275	280
1.21	0.5324	0.5172	0.5072	0.4637	0.4560	0.4292
1.23	0.5375	0.5176	0.4942	0.4671	0.4570	0.4287
1.25	0.5426	0.5212	0.4974	0.4665	0.4539	0.4326
1.27	0.5436	0.5238	0.5032	0.4699	0.4474	0.4265
1.29	0.5553	0.5324	0.5064	0.4731	0.4539	0.4339
1.31	0.5726	0.5559	0.5199	0.4881	0.4609	0.4334
1.34	0.5910	0.5586	0.5281	0.4819	0.4652	0.4388
1.37	0.6168	0.5848	0.5455	0.5026	0.4742	0.4525
1.40	0.6289	0.6005	0.5634	0.5213	0.4864	0.4527
1.44	0.6592	0.6159	0.5751	0.5305	0.4988	0.4629
1.47	0.6767	0.6473	0.5974	0.5333	0.5056	0.4598
1.50	0.7229	0.6720	0.6190	0.5675	0.5288	0.4922
1.55	0.7581	0.6918	0.6397	0.5925	0.5493	0.4988
1.59	0.7972	0.7278	0.6626	0.6038	0.5584	0.5159
1.64	0.8326	0.7562	0.7005	0.6410	0.5810	0.5259
1.69	0.8544	0.7758	0.7073	0.6385	0.5842	0.5368
1.75	0.8876	0.8073	0.7377	0.6654	0.6034	0.5530
1.83	0.9332	0.8427	0.7584	0.6757	0.6236	0.5698
1.90	0.9521	0.8657	0.7782	0.6993	0.6297	0.5621
2.01	0.9799	0.8863	0.8004	0.7069	0.6435	0.5796
2.09	0.9830	0.8932	0.8003	0.7151	0.6389	0.5806
2.19	1.0075	0.9002	0.8034	0.7212	0.6499	0.5885
2.32	1.0081	0.9018	0.8096	0.7154	0.6519	0.5914
2.50	1.0021	0.8958	0.8117	0.7267	0.6522	0.5907

Table A5. Experimental Values of Absorbance at Different pH and Wavelengths at T = 298 K, I = 1.05 mol·kg^{-1}

	λ/nm					
pH	255	260	265	270	275	280
1.09	0.6449	0.6180	0.6036	0.5844	0.5660	0.5408
1.16	0.6165	0.5913	0.5686	0.5649	0.5396	0.5075
1.20	0.5971	0.5746	0.5444	0.5427	0.5218	0.4920
1.22	0.5876	0.5545	0.5413	0.5340	0.5052	0.4782
1.24	0.5858	0.5460	0.5334	0.5090	0.4990	0.4798
1.26	0.5735	0.5484	0.5295	0.5131	0.4897	0.4719
1.27	0.5649	0.5341	0.5138	0.5040	0.4715	0.4573
1.28	0.5702	0.5471	0.5201	0.5049	0.4683	0.4496
1.29	0.5897	0.5500	0.5294	0.5011	0.4769	0.4464
1.30	0.6272	0.5780	0.5394	0.5122	0.4832	0.4507
1.31	0.6389	0.5978	0.5561	0.5234	0.4923	0.4605
1.34	0.6689	0.6090	0.5807	0.5391	0.4960	0.4600
1.37	0.6951	0.6379	0.6046	0.5473	0.5036	0.4723
1.41	0.7394	0.6744	0.6120	0.5777	0.5251	0.4770
1.46	0.7796	0.7121	0.6422	0.5981	0.5409	0.4985
1.51	0.7424	0.6817	0.6241	0.5634	0.5144	0.5183
1.56	0.8774	0.7854	0.7138	0.6396	0.5851	0.5245
1.62	0.9138	0.8125	0.7337	0.6663	0.6027	0.5539
1.70	0.9419	0.8646	0.7779	0.7076	0.6328	0.5463
1.79	0.9899	0.8862	0.7849	0.7143	0.6384	0.5790
1.88	1.0226	0.8972	0.8034	0.7218	0.6540	0.5869
2.02	1.0435	0.9256	0.8329	0.7453	0.6638	0.5958

Table A5. Continued

pH	λ/nm					
	255	260	265	270	275	280
2.13	1.0600	0.9369	0.8388	0.7499	0.6634	0.5977
2.28	1.0496	0.9547	0.8514	0.7504	0.6657	0.6055
2.50	1.0617	0.9502	0.8356	0.7567	0.6649	0.5882

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