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A Potentiometric and Polarimetric Determination of the Stability Constants of the Cobalt(II)—(+)-Lactate Complexes in Aqueous Solution

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The complexity of the cobalt(II)–(+)-lactate system has been investigated by a polarimetric and a potentiometric method. Both methods yield values of the stability constants (*cf.* Table 8) that are in good agreement within the experimental errors.

The molar rotation of a metal ion complex containing an optically active ligand has been postulated [1] to be as characteristic of the complex as is the molar absorption coefficient. Consequently, the same methods may be applied for the elucidation of complex equilibria by using optical rotation measurements as those that are used in connection with ordinary light absorption measurements. Such investigations have been reported recently from this laboratory.[1,2]

In order to further substantiate the above mentioned postulate we have now also investigated the cobalt(II)–(+)-lactate system. This system was chosen also with the aim of further studying [3] the circular dichroism of cobalt(II) chelate systems, one such investigation having been reported recently.[4] A conventional potentiometric method has been applied to check the results of the polarimetric investigation.

The presentation of the investigations will follow in detail the earlier papers,[1,2] where any symbols or notations not defined in the present report will be found explained.

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THE POLARIMETRIC INVESTIGATION

As it was desirable that the rotation of the free lactic acid should enter into the analytical expressions as a small correction only, we used buffers of lactate and lactic acid with a concentration ratio of 400 : 1. The pH of such solutions is about 6 and hence the influence of hydrolytic products is negligible.[5] As before,[1,2] the ionic strength was kept constant (2 M) by using sodium perchorate as supporting electrolyte.

The solutions, the optical rotations of which were measured, thus had the general composition:

$$\begin{array}{ll} C_{L}' \text{ M} & \text{NaL} \\ C_{HL} \text{ M} & \text{HL} \\ C_{M} \text{ M} & \text{Co}(\text{ClO}_4)_2 \\ (2 - C_{L}' - 3C_{M}) \text{ M} & \text{NaClO}_4 \end{array}$$

Once the molar rotations, δ_{L} and δ_{HL}, of lactate ion and lactic acid, respectively, are measured, one can easily calculate the function ϕ from the observed angle of rotation, θ', (*cf.* eqns. (1) and (3) of Ref. 1).

Now assuming that no polynuclear complexes are formed under the experimental conditions, ϕ has the form

$$\phi = X^{-1} \sum_{n=1}^{N} \delta_n \beta_n [\text{L}]^n - \delta_{L} \bar{n} \qquad (1)$$

Here δ_n means the molar rotation of the complex ML_n. Thus, as ϕ is dependent only on [L], one can obtain the mean ligand number $\bar{n}$ as a function of free ligand [L]. (For details, see Ref. 1.) Consequently [6] the stability constants can be calculated graphically from the expression.

$$\ln X([\text{L}]_i) = \int_{0}^{[\text{L}]_i} \frac{\bar{n}}{[\text{L}]} \, \text{d}[\text{L}] \qquad (2)$$

Experimental

The measurements were made with a Perkin Elmer Model 141 photoelectric polarimeter. Each reading (to 0.001°) was repeated at least four times. The values reported in Table 3 or otherwise used for calculations are the corresponding mean values. The 10 cm cells were thermostated to 20.0°C. With the help of interference filters the following lines from a mercury lamp source were isolated: 578, 546, 436 and 365 nm. The measurements were performed at these four wavelengths.

Chemicals. Cobalt(II) perchlorate was prepared from analytical grade cobalt(II) carbonate and perchloric acid. A stock solution was prepared from the recrystallized salt. The cobalt concentration was determined by EDTA titration (following Harris [7]) to an accuracy better than 0.1 %. The excess of free acid was found to be 0.279 % of the cobalt concentration (cationic exchange – alkalimetric titration).

A lactic acid stock solution was prepared from crystalline L(+)-lactic acid (Sigma Chemical Co.) and the concentration was determined by a potentiometric titration using the graphical treatment of Gran.[8] Sodium perchlorate and perchloric acid were of the same origin as before.[1]

Measurements

The determination of the optical activity of a solution of L(+)-lactic acid is a difficult task [9] as the rotation seems to depend on concentration and varies with time. Bancroft and Davis [10] have shown, in fact, that solutions of lactic acid may contain three laevorotatory forms (the normal, anhydride and lactide ones) and one dextrorotatory form (an epoxy type). The positions of equilibrium of these different forms are reached with different velocities depending upon dilution, so the actual rotation of a solution depends on concentration and time. In order to avoid this effect in the present investigation we have chosen to work with as low concentrations as possible, *i.e.* $C_{HL}' \leq 0.001$ M and $C_{L}' \geq 0.400$ M. From the work of Davis [10] it then follows that the molar rotation of the species of the buffer would be constant. Indeed, during a time span of two months no alteration of the optical rotation of the buffer solutions was observed.

In order to check that there was no complex formation between the lactic acid and the metal ion, we measured the rotation of solutions with or without ($C_M = 0$) cobalt but otherwise of equal composition, C_{HL} M HL, C_M M $Co(ClO_4)_2$, $(2 - 3C_M)$ M $HClO_4$. In both cases, δ_{HL} was found to be practically constant. Therefore, we conclude that lactic acid does not form any complexes with Co(II) ions.

Table 1. Molar rotation of lactic acid.

λ nm	578	546	436	365
δ_{HL} degrees M^{-1} dm^{-1}	0.583	0.693	1.476	3.025

Table 2. Molar rotation of lactate ions as a function of C_L. $I = 2$ M.

C_L mM	$-\delta_L$ degrees M^{-1} dm^{-1}			
	578 nm	546 nm	436 nm	365 nm
25	1.441	1.602	2.644	3.887
50	1.401	1.562	2.564	3.787
100	1.361	1.532	2.523	2.727
150	1.341	1.522	2.497	3.681
200	1.336	1.517	2.488	3.667
300	1.325	1.501	2.473	3.647
400	1.311	1.487	2.456	3.622

Table 3. Survey of the polarimetric measurements. Concentrations are given in mM, θ' in degrees, and ϕ in degrees M^{-1} dm^{-1}.

$\lambda = 436$ nm, $C_M = 15$ mM
C_L, $-\theta'$, ϕ: 18.33, −0.002, 3.33; 24.58, 0.002, 4.13; 37.09, 0.009, 5.73; 49.58, 0.014, 7.47; 74.58, 0.033, 10.33; 84.58, 0.041, 11.47; 99.58, 0.064, 12.40; 112.09, 0.078. 13.53; 124.58, 0.094, 14.47; 149.58, 0.133, 16.00; 169.58, 0.163, 17.27; 199.58. 0.212, 18.87; 234.58, 0.272, 20.67; 269.58, 0.340, 21.67; 304.58, 0.402, 23.20; 339.58, 0.470, 24.33.

Table 3. Continued.

$\lambda = 436$ nm, $C_M = 25$ mM
C_L, θ', ϕ: 18.05, 0.023, 2.80; 24.30, 0.027, 3.60; 36.80, 0.040, 5.36; 49.30, 0.046, 6.88; 74.30, 0.050, 9.56; 86.80, 0.049, 10.72; 99.30, 0.044, 11.72; 111.80, 0.038, 12.72; 124.30, 0.031, 13.68; 149.30, 0.013, 15.40; 174.30, −0.013, 16.76; 199.30, −0.043, 18.04; 224.30, −0.072, 19.32; 249.30, −0.105, 20.44; 274.30, −0.141, 21.44; 299.30, −0.178, 22.40.

$\lambda = 436$ nm, $C_M = 50$ mM
C_L, θ', ϕ: 17.35, 0.059, 2.08; 23.60, 0.077, 2.74; 36.10, 0.111, 4.06; 48.60, 0.140, 5.26; 73.60, 0.194, 7.58; 86.10, 0.214, 8.60; 98.60, 0.234, 9.62; 111.10, 0.253, 10.60; 123.60, 0.267, 11.50; 148.60, 0.290, 13.16; 173.60, 0.305, 14.70; 198.60, 0.312, 16.06; 223.60, 0.315, 17.34; 248.60, 0.310, 18.46; 273.60, 0.301, 19.50; 298.60, 0.286, 20.42.

$\lambda = 365$ nm, $C_M = 15$ mM
C_L, $-\theta'$, ϕ: 18.33, 0.039, 2.13; 24.58, 0.055, 2.67; 37.09, 0.083, 3.87; 49.58, 0.112, 4.93; 74.58, 0.173, 7.00; 84.58, 0.195, 7.93; 99.58, 0.243, 8.40; 112.09, 0.275, 9.40; 124.58, 0.309 10.00; 149.58, 0.383, 11.07; 169.58, 0.439, 12.20; 199.58, 0.529, 13.33; 234.58, 0.637, 14.60; 269,58, 0.747, 15.60; 304.58, 0.854, 16.87; 339.58, 0.966, 17.67.

$\lambda = 365$ nm, $C_M = 25$ mM,
C_L, $-\theta'$, ϕ: 18.05, 0.023, 1.92; 24.30, 0.036, 2.28; 36.80, 0.049, 3.60; 49.30, 0.070, 4.60; 74.30, 0.113, 6.52; 86.80, 0.138, 7.32; 99.30, 0.166, 8.04; 111.80, 0.192, 8.80; 124.30, 0.220, 9.44; 149.30, 0.279, 10.72; 174.30, 0.342, 11.84; 199.30, 0.410, 12.68; 224.30, 0.474, 13.72; 249.30, 0.542, 14.64; 274.30, 0.612, 15.40; 299.30, 0.682, 16.20.

$\lambda = 365$ nm $C_M = 50$ mM,
C_L, $-\theta'$, ϕ: 17.35, −0.003, 1.36; 23.60, 0.000, 1.76; 36.10, 0.002, 2.64; 48.60, 0.008, 3.42; 73.60, 0.015, 5.12; 86.10, 0.027, 5.80; 98.60, 0.037, 6.50; 111.10, 0.047, 7.20; 123.60, 0.058, 7.86; 148.60, 0.087, 9.10; 173.60, 0.119, 10.28; 198.60, 0.161, 11.22; 223.60, 0.202, 12.22; 248.60, 0.253, 13.00; 273.60, 0.298, 13.90; 298.60, 0.350, 14.66.

Table 4. Calculations of the stability constants from the polarimetric measurements. The following values were obtained: $\beta_1 = 24 \pm 1$ M^{-1}, $\beta_2 = 200 \pm 30$ M^{-2}, $\beta_3 = 200 \pm 100$ M^{-3}.

[L] mM	$\bar{n}$ $\lambda = 436$ nm	$\bar{n}$ $\lambda = 365$ nm	$\bar{n}$ mean	X	X M^{-1}	X_2 M^{-2}	X_3 M^{-3}
5	0.112	0.108	0.110	1.12	24.8		
10	0.220	0.200	0.210	1.26	26.0		
15	0.330	0.290	0.310	1.41	27.0		
20	0.430	0.380	0.405	1.56	28.0	205	
25	0.520	0.467	0.500	1.73	29.1	207	288
30	0.600	0.548	0.576	1.90	30.1	208	258
35	0.680	0.632	0.654	2.09	31.2	209	268
40	0.750	0.710	0.730	2.29	32.3	209	230
50	0.880	0.834	0.860	2.73	34.6	213	264
60	1.030	0.937	0.970	3.21	36.9	216	263
70	1.080	1.020	1.050	3.74	39.1	217	244
80	1.130	1.092	1.110	4.31	41.4	218	230
90	1.168	1.168	1.168	4.93	43.7	220	219
100	1.200	1.237	1.218	5.59	45.9	220	204
120	1.290	1.375	1.330	7.06	50.5	222	182
140	1.435	1.492	1.463	8.74	55.3	224	171
160	1.590	1.595	1.592	10.70	60.6	230	184
180	1.730	1.676	1.703	13.00	66.7	238	209

The values of δ_{HL} determined at the four different wavelengths are given in Table 1. They were determined for $C_{HL} \leqslant 0.100$ M. (For higher concentrations deviation from constancy was observed.)

The molar rotation, δ_L, of the lactate ion is given in Table 2. As there is a slight variation with concentration, we used graphical interpolation from Table 2 to determine the values used in the calculation of ϕ for each value of C_L.

The results of the main measurements are given in Table 3. In each series of measurements C_M is kept constant ($C_M = 10$, 25, and 50 mM). Because of the strong light absorption of Co(II) at 546 nm it was not possible to obtain any readings at all for $C_M = 50$ mM. Therefore, readings for $C_M = 10$ and 25 mM at this wavelength could not be utilized. Furthermore, also at $\lambda = 578$ nm the accuracy of the readings was not sufficiently high, probably because of the combined effect of high absorption and a low response of the photo multiplier. Therefore, we have chosen to report only those readings that were actually used in the calculation of $\bar{n}$, *i.e.* at $\lambda = 436$ and 365 nm.

Results

The calculations leading to the stability constants are given in Table 4. It is observed that three complexes exist within the concentration range investigated.

THE POTENTIOMETRIC INVESTIGATION

The method is based on the determination of the free ligand concentration *via* the measurement of the hydrogen ion concentration with a glass electrode. With no metal salt present the following expression holds:

$$K_a = \frac{[H^+]'(C_L' + [H^+]')}{C_{HL}' - [H^+]'} \tag{3}$$

When some metal salt of concentration C_M is also present, the measured hydrogen ion concentration is $[H^+]$ and

$$K = \frac{[H^+]\,[L]}{C_{HL} + C_H - [H^+]} \tag{4}$$

C_H is the concentration of free perchloric acid that is present in the solution when the metal perchlorate concentration is C_M.

Combining (3) and (4) we obtain:

$$[L] = \frac{[H^+]'(C_L' + [H^+]')\,(C_{HL}' + C_H - [H^+])}{[H^+]\,(C_{HL}' - [H^+])} \tag{5}$$

and hence the mean ligand number $\bar{n}$:

$$\bar{n} = \frac{C_L' - C_H + [H^+] - [L]}{C_M} \tag{6}$$

From the ($\bar{n}$, [L]) values thus obtained the calculations of the stability constants were performed as above (*cf.* eqn. (2)).

Experimental

The emf, E, of galvanic cells of the following composition was measured

– Ag, AgCl	10.0 mM NaCl I = 2 M ($NaClO_4$)	2 M $NaClO_4$	C_M M $Co(ClO_4)_2$ C_H M $HClO_4$ C_L' M NaL C_{HL}' M HL (2-C_L' – 3C_M) M $NaClO_4$	glass electrode+

As usual, the composition of the right half-cell was obtained from a titration procedure. A stream of oxygen-free nitrogen was passed through the solution. Two different buffer solutions were used, *viz.*, $C'_L/C'_{HL} = 1:1$ and $3:1$, respectively. The total metal perchlorate concentration was kept constant during each titration. Three values were used, $C_M = 10$, 25, and 50 mM. Titrations were also performed at $C_M = 0$. The emf in this case is denoted by E'. The ratio $[H^+]/[H^+]'$ could thus be calculated from the relation

$$E - E' = 58.21 \log [H^+]/[H^+]'$$

In order to get absolute values of $[H^+]$ and $[H^+]'$ we measured the emf, E'', of a cell in which the right half-cell had the composition $[H^+]'' = 10.0$ mM $HClO_4$; $I = 2$ M ($NaClO_4$).

Hence from the expression

$$E'' - E = 58.21 \log [H^+]''/[H] \qquad (7)$$

we could obtain $[H^+]$ (or $[H^+]'$, respectively).

Every titration was repeated at least twice and the reproducibility of the potential values, referred to the same E_0, was ± 0.2 mV. Before and after each titration it was checked that the asymmetry potential of the glass electrode had not changed.

All emf measurements were performed with a Radiometer pH Meter Type 4 e and glass electrodes of Radiometer Type G 202 B. The coefficient in the Nernst expression was determined to be 58.21 mV. The Ag/AgCl electrode was prepared according to Brown.[11]

The temperature of the solutions was kept at 20.00 ± 0.05°C.[1] The chemicals were the same as used in the polarimetric work.

Results

The values of E' determined for the two buffers are given in Table 5. Table 6 presents the main material, E for each combination of C_L', buffer composition and C_M, together with the resulting values of [L] and $\bar{n}$. From these, $\bar{n}$ was calculated and plotted against [L] (*cf.* Fig. 1). It is immediately seen that $\bar{n}$ does not depend upon the buffer composition (*i.e.*, as the low pH makes hydrolytic products improbable [5] one can conclude that no acidic complexes are formed) nor upon C_M (*i.e.* no polynuclear complexes are formed). Hence from a smoothed curve through the experimental points $\bar{n}/[L]$ the integration to give X (eqn. (2)) was performed. Table 7 gives the details of the subsequent calculation of the stability constants as well as a comparison between the (smoothed) experimental and the calculated values of $\bar{n}/[L]$. As is also seen from Fig. 1 the stability constants thus derived describe the system well.

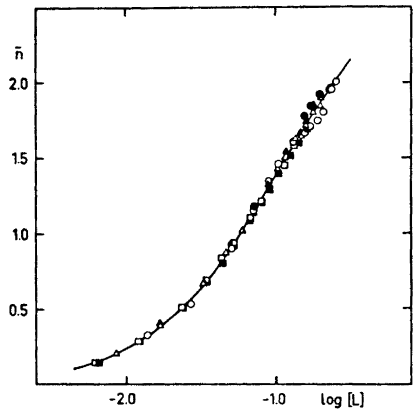

Fig. 1. $\bar{n}$ calculated from the potentiometric measurements. Filled points refer to buffer 1 : 3, unfilled ones to buffer 1 : 1. $C_M = 10$ nM, ○; $C_M = 25$ nM, △; and $C_M = 50$ nM, □.

Table. 5. Determination of E' as a function of $C_{L'}$. The values are referred to $E'' = 160.8$ mV.

$C_{L'}$ mM	Buffer $C_{L'}/C_{HL'} = 1:1$ $pK_{L'} = 3.80 \pm 0.02$	Buffer $C_{L'}/C_{HL'} = 1:3$ $pK_{L'} = 3.77 \pm 0.02$
	E' mV	E' mV
5.45	50.9	12.9
10.00	54.8	22.6
13.84	56.3	25.5
17.14	56.9	26.9
20.00	57.2	27.7
24.71	57.8	28.7
28.42	58.0	29.1
31.43	58.1	29.4
35.00	58.3	29.8
37.78	58.5	30.0
40.64	58.5	30.1
47.86	58.4	30.2
66.66	57.9	30.6
133.33	57.8	30.5
200.00	57.2	30.1
240.00	57.1	29.9
266.70	57.0	29.7
285.71	56.8	29.6
300.00	56.7	29.5
320.00	56.6	29.4
333.33	56.6	29.3

Table 6. Determination of corresponding values of [L] and $\bar{n}$ from the measured emf.

Buffer HL : L = 1 : 1											
C_M = 10 mM				C_M = 25 mM				C_M = 50 mM			
C_L' mM	E mV	[L] mM	$\bar{n}$	C_L' mM	E mV	[L] mM	$\bar{n}$	C_L' mM	E mV	[L] mM	$\bar{n}$
17.1	62.7	14.0	0.312	14.3	70.0	8.7	0.206	14.3	79.9	6.1	0.144
32.7	63.0	27.4	0.52	27.3	70.2	17.2	0.385	27.3	79.5	12.2	0.282
60.0	62.7	51.0	0.90	50.0	69.3	33.1	0.66	50.0	78.2	23.6	0.51
83.1	62.2	71.6	1.14	69.2	68.5	47.0	0.87	69.2	77.0	33.8	0.69
102.9	61.8	89.3	1.35	85.7	67.7	59.9	1.02	85.7	75.9	43.6	0.82
120.0	61.5	105.7	1.46	100.0	67.2	70.9	1.15	100.0	75.0	52.4	0.93
148.2	61.0	132.5	1.60	123.5	66.4	89.9	1.33	123.5	73.8	67.4	1.10
170.5	60.6	154.2	1.66	142.1	65.7	105.8	1.43	142.1	72.8	80.6	1.21
188.6	60.3	171.9	1.70	157.1	65.2	118.8	1.52	157.1	72.1	90.8	1.31
210.0	60.0	192.9	1.74	175.0	64.8	133.8	1.63	175.0	71.2	104.2	1.39
226.7	59.8	209.0	1.80	188.9	64.3	147.3	1.65	188.9	70.6	115.2	1.45
243.9	59.6	225.7	1.85	203.2	64.0	159.7	1.72	203.2	70.2	125.3	1.54
257.1	59.5	238.0	1.95	214.3	63.7	169.7	1.77	214.3	69.8	133.7	1.58
270.0	59.4	249.8	2.05	225.0	63.5	179.5	1.80	225.0	69.4	142.5	1.63
288.0	59.1	268.6	1.97	240.0	63.2	193.0	1.86	240.0	68.7	156.3	1.65
300.0	59.0	279.8	2.05	250.0	62.8	203.4	1.84	250.0	68.4	163.4	1.71
Buffer HL : L = 1 : 3											
17.1	34.0	13.8	0.368	14.3	41.7	8.8	0.219	14.3	53.3	6.7	0.138
32.7	35.4	26.8	0.63	27.3	42.5	17.1	0.405	27.3	52.5	12.5	0.281
60.0	35.2	50.9	0.94	50.0	41.8	33.1	0.68	50.0	51.1	24.1	0.50
83.1	34.9	71.6	1.18	69.2	41.0	47.3	0.87	69.2	49.8	34.7	0.68
102.9	34.5	89.8	1.34	85.7	40.3	60.2	1.02	85.7	48.8	44.8	0.80
120.0	34.1	105.9	1.44	100.0	39.7	74.5	1.02	100.0	47.9	53.2	0.92
148.2	33.7	131.6	1.69	123.5	38.9	90.1	1.34	123.5	46.5	68.0	1.09
170.5	33.3	153.2	1.77	142.1	38.3	105.9	1.44	142.1	45.5	81.1	1.21
188.6	33.0	170.5	1.84	157.1	37.8	118.4	1.55	157.1	44.7	91.9	1.29
210.0	32.7	191.4	1.92	175.0	37.2	134.8	1.60	175.0	43.9	104.9	1.39
226.7	32.5	207.3	1.93	188.9	36.9	147.1	1.67	188.9	43.3	115.7	1.45
243.9	32.2	224.7	1.95	203.2	36.6	159.5	1.75	203.2	42.7	126.8	1.51
257.1	32.0	237.8	1.97	214.3	36.3	169.3	1.79	214.3	42.2	135.6	1.56
270.0	31.7	251.6	1.87	225.0	36.1	179.2	1.86	225.0	41.8	144.6	1.59
288.0	31.5	269.4	1.89	240.0	35.7	193.3	1.86	240.0	41.4	155.9	1.67
300.0	31.3	281.7	1.86	250.0	35.5	202.1	1.91	250.0	41.0	164.1	1.70

DISCUSSION

Table 8 presents a comparison between the results obtained with the two methods. It is seen that a good agreement exists for the first and also the second stability constant. Some deviation occurs for the third complex but considering that the limits of error are of the same order of magnitude as the measured quantity, the discrepancy is not large.

Both methods most clearly indicate that only three complexes are present. Thus the existence of a fourth complex claimed by Calabro and Cuzzo [12] seems very doubtful.

Table 7. Calculation of the stability constants from the potentiometric measurements. The following values were obtained: $\beta_1 = 24.5 \pm 1.0$ M^{-1}; $\beta_2 = 230 \pm 20$ M^{-2}; $\beta_3 = 550 \pm 200$ M^{-3}.

[L] mM	$\bar{n}$/[L] M^{-1}	X	X_1 M^{-1}	X_2 M^{-2}	X_3 M^{-3}	$\bar{n}$/[L] calc.
5	23.9	1.13	25.8			23.8
10	23.1	1.27	27.0			23.1
15	22.4	1.42	28.2			22.4
20	21.7	1.59	29.5			21.7
25	21.0	1.77	30.7	249		21.0
30	20.3	1.96	32.0	250		20.4
35	19.7	2.17	33.3	252		19.8
40	19.1	2.39	34.7	254	555	19.2
50	18.5	2.87	37.5	259	544	18.4
60	17.0	3.42	40.4	265	542	17.0
70	16.1	4.04	43.4	270	541	16.1
80	15.3	4.72	46.5	276	544	15.3
90	14.7	5.49	49.9	282	552	14.5
100	13.9	6.33	53.3	288	562	13.8
120	12.6	8.24	60.4	299	558	12.6
140	11.6	10.49	67.8	309	551	11.6
160	10.7	13.12	75.8	320	552	10.8
180	10.1	16.15	84.2	332	553	10.0
200	9.3	19.61	93.1	343	554	9.4
220	8.6	23.44	102.0	352	547	8.8

Table 8. Comparison between the values of β_n (M^{-n}) and K_n (M^{-1}) determined polarimetrically and potentiometrically. In calculating the ratios K_n/K_{n+1}, the mean values of K_n have been used.

Polarography		Potentiometry		K_n/K_{n+1}
$\beta_1 = 24 \pm 1$	$K_1 = 24$	$\beta_1 = 24.5 \pm 1.0$	$K_1 = 24.5$	
$\beta_2 = 200 \pm 30$	$K_2 = 8.3$	$\beta_2 = 230 \pm 20$	$K_2 = 9.4$	$K_1/K_2 = 2.8$
$\beta_3 = 200 \pm 100$	$K_3 = 1$	$\beta_3 = 550 \pm 200$	$K_3 = 2$	$K_2/K_3 = 5.8$

The fact that only three complexes are formed could be taken as an indication of a preferentially chelated type of bonding, as in such a case all six coordination sites of the central atom should be occupied. However, the ratios between the formation constants K_n/K_{n+1} given in Table 8 seem to indicate no great difference from what is found for monodentate systems. A full discussion of these problems will, however, be postponed until a later publication.[3]

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