CHELATE-FORMING PROPERTIES OF N,N-BIS(CARBOXYMETHYL)AMINOACETOHYDROXAMIC ACID

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Preparation, acid-base and chelate-forming properties of a new reagent with one hydroxamic functional group derived from nitrilotriacetic acid are described. Formation of chelates with Cu(II): CuLH, CuL^-, $CuL(OH)^{2-}$, $CuL(OH)_2^{3-}$ and with Fe(III): $FeLH^+$, FeL, $FeL(OH)^-$, $FeL(OH)_2^{2-}$ has been established spectrophotometrically. Equilibrium and stability constants of the individual chelates are given along with their probable structure.

The present work represents a continuation in the study of analogs of aminopolycarboxylic acids with hydroxamic functional group as chelate-forming reagents[1]. It deals with preparation and properties of a reagent derived from nitrilotriacetic acid having one carboxylic group substituted by hydroxamic group.

EXPERIMENTAL

Reagents and Apparatus

The stock solutions 10^{-2}M-$Cu(ClO_4)_2$ and $Fe(ClO_4)_3$ were prepared from the respective perchlorates (Fluka) and their concentration was determined chelatometrically with murexide resp. sulfosalicylic acid as the indicators. pH of the solutions was adjusted with carbonate-free 0·1M and 1M-NaOH resp 0·1M and 1M-$HClO_4$ added by means of a plunger microburette ABU-12 (Radiometer). Ionic strength I 0·1 was adjusted with 1M-$NaClO_4$ in all the solutions except for those of pH < 1. The spectrophotometric measurements were carried out with a Unicam SP 1700 spectrophotometer and a Perkin Elmer 577 infrared spectrophotometer. pH of the solutions was determined with a PHM-52 apparatus (Radiometer) using the combined GK 2321C electrode (Radiometer).

Synthesis of N,N-Bis(carboxymethyl)aminoacetohydroxamic Acid

The preparation of the title substance started from iminodiacetic acid and ethyl monobromoacetate; the ester obtained reacted with hydroxylamine in methanol.

13·4 g (0·1 mol) iminodiacetic acid was dissolved in 50 ml water and 30 g (0·3 mol) potassium hydrogen carbonate and 50 ml ethanol were added successively, whereupon the mixture was heated to 80°C with continuous stirring. During one hour 11·1 ml (0·1 mol) ethyl bromoacetate was added, and the clear solution was maintained at 80°C for further 3 hours. After cooling and

concentrating to sirupy consistency, 50 ml methanol was added to remove KBr. Methanolic solution of the ester was passed through a column filled with Amberlite IRC-50 exchanger (H-cycle), using methanol as eluent. The main portion was concentrated in vacuum and left for crystallization. Ethyl potassium hydrogen nitrilotriacetate obtained in this way was very pure (min 99%). Yield 15·5 g (60%). For $C_8H_{12}KNO_6$ (258·3) calculated: 37·20% C, 4·68% H, 5·42% N; found: 37·15% C, 4·80% H, 5·40% N.

2·6 g (10 mmol) Ethyl potassium hydrogen nitrilotriacetate was dissolved in 10 ml 4M methanolic hydroxylamine with stirring, and the solution was stirred in a closed flask at the room temperature for 4 hours. Then 10 ml 1M potassium methoxide was added dropwise and the mixture was stirred for 8 hours. The reaction mixture was treated with 50 ml ethanol, and the precipitated solid was filtered off and washed with ethanol. Yield 2·1 g (77%) raw dipotassium salt of N,N-bis-(carboxymethyl)aminoacetohydroxamic acid. The substance obtained can be purified by repeated precipitation, *i.e.* dissolution in methanol, filtration off of the non-dissolved portions, and addition of the same volume of ethanol. The product forms fine colourless crystals stable in air, melting at 198 to 200°C with decomposition. It is very well soluble in methanol, less in ethanol, and insoluble in ether. For $C_6H_8K_2N_2O_6$ (282·3) calculated: 25·53% C, 2·85% H, 9·92% N; found: 25·21% C, 2·93% H, 9·89% N.

Determination of Dissociation Constants

Solutions of the reagent of the concentrations (0·7 to 1·2) . 10^{-3}M were prepared for each measurement in redistilled water, and the ionic strength I 0·1 was adjusted. The titrations were carried out in a jacketed thermostatted vessel at 20 ± 0·2°C with carbonate-free 0·1M-NaOH resp. 0·1M-$HClO_4$ with stirring with a stream of nitrogen. Before each measurement the pH meter was calibrated with standard buffer solutions: potassium hydrogen phthalate (pH 4·002), phosphate buffer (pH 7·429) and borax buffer (pH 9·225). For determination of the dissociation constant K_{a1} the titrations were carried out with more concentrated solutions of the reagent (10^{-2}M). Calculation of the constants from individual buffer regions was carried out with a computer according to the program POT-3 (ref.[2]).

In the spectrophotometric determination of the dissociation constant the absorbance of the reagent solutions (5·2 . 10^{-5} to 5·6 . 10^{-5}M) was measured at 216 and 230 nm in 100 ml thermostatted quartz cells with simultaneous measurement of pH and stirring with a stream of nitrogen[1]. Calculation was carried out in the usual way[3].

Spectrophotometric Investigation of the Complexes in Solution

Formation of the individual complexes was followed by measuring the absorption spectra of the solutions with various excesses of the reagent in a broad pH range. Very detailed investigation was allowed by adaptation of the spectrophotometer[1] enabling gradual pH adjusting and simultaneous precise absorbance and pH measurements. The A–pH curves (c_M, c_L = const.) thus obtained were subjected to graphical and logarithmic analysis[4] wherefrom the existence of individual equilibria in the solution was determined along with molar absorption coefficients and equilibrium and stability constants of the individual complexes. Composition of the complexes was determined by graphical analysis of the absorption curves of the

solutions with varying concentration of the reagent (pH, c_M = const.) and of the Job curves of the isomolar solutions.

Basic equations and the corresponding transformations which are to be considered for formation of complexes of the reagent studied[4]:

1. Equilibrium with the complex formation (written without the charges):

$$M + n\,H_xL \rightleftharpoons ML_nH_z + q\,H^+ \qquad (A)$$

$$^*K = [ML_nH_z][H]^q/[M][H_xL]^n \qquad (1)$$

For solutions with excess of the reagent:

$$c_M/(A - \varepsilon_M c_M) = 1/(\varepsilon_n - \varepsilon_M) + [H]^q/(\varepsilon_n - \varepsilon_M)c_L^n\,^*K \qquad (2)$$

$$A = \varepsilon_n c_M - [H]^q\,(A - \varepsilon_M c_M)/c_L^n\,^*K \qquad (3)$$

$$\log((A - \varepsilon_M c_M)/(\varepsilon_n c_M - A)) = n \log c_L + q\,pH + \log\,^*K \qquad (4)$$

For the equimolar solutions:

$$A = \varepsilon_n c_M - ((A - \varepsilon_M c_M)[H]^q\,(\varepsilon_n - \varepsilon_M)/^*K)^{1/2} \qquad (5)$$

$$\log((A - \varepsilon_M c_M)/(\varepsilon_n c_M - A)^2) = q\,pH - \log(\varepsilon_n - \varepsilon_M) + \log\,^*K \qquad (6)$$

2. Dissociation of the protonated complex:

$$ML_nH_z \rightleftharpoons ML_nH_{z-q} + q\,H^+ \qquad (B)$$

$$^*K' = [ML_nH_{z-q}][H]^q/[ML_nH_z] \qquad (7)$$

$$c_M/A = 1/\varepsilon_2 + [H]^q\,(A - \varepsilon_1 c_M)/A\varepsilon_2\,^*K' \qquad (8)$$

$$A = \varepsilon_2 c_M - (A - \varepsilon_1 c_M)[H]^q/^*K' \qquad (9)$$

$$\log((A - \varepsilon_1 c_M)/(\varepsilon_2 c_M - A)) = q\,pH + \log\,^*K' \qquad (10)$$

3. Hydrolysis of the complex:

$$ML + q\,H_2O \rightleftharpoons ML(OH)_q + q\,H^+ \qquad (C)$$

$$k = [ML(OH)_q][H]^q/[ML] \qquad (11)$$

$$c_M/A = 1/\varepsilon_2 + (A - \varepsilon_1 c_M)\,[H]^q/A\varepsilon_2 k \qquad (12)$$

$$A = \varepsilon_2 c_M - (A - \varepsilon_1 c_M)\,[H]^q/k \qquad (13)$$

$$\log((A - \varepsilon_1 c_M)/(\varepsilon_2 c_M - A)) = q\ \mathrm{pH} + \log k \qquad (14)$$

RESULTS AND DISCUSSION

N,N-*Bis*(*carboxymethyl*)*aminoacetohydroxamic Acid*

Potenciometric titration curve of dipotassium salt of the ligand shows a marked potential break for 1 equivalent of acid per 1 mol of ligand, and a less distinct break for 1 equivalent of base per mol of ligand. Potentiometric determination of the dissociation constants was carried out both with freshly prepared solutions and with the old ones (several hours to 30 days). In all the cases the obtained values of dissociation constants agreed, which among others indicates stability of the studied reagent

TABLE I
Summary of Individual Constants
I 0·1 ($NaClO_4$), 20°C.

Equilibrium	Constant	Log of the constant
$[H_2L]\,[H]/[H_3L]$	K_{a1} (—COOH)	$-2{\cdot}44 \pm 0{\cdot}01$
$[HL]\,[H]/[H_2L]$	K_{a2} (—NH^+)	$-6{\cdot}16 \pm 0{\cdot}01$
$[L]\,[H]/[HL]$	K_{a3} (—CONHOH)	$-9{\cdot}42 \pm 0{\cdot}02$
		$-9{\cdot}35 \pm 0{\cdot}03^a$
$[CuLH]\,[H]^2/[Cu]\,[H_3L]$	$^*K_{12}$	0·85; 0·88[b]
$[CuLH]/[Cu]\,[HL]$	β_{1H}	9·45
$[CuL]\,[H]/[CuLH]$	$^*K'$	−4·15
$[CuL]/[Cu]\,[L]$	β_1	14·72
$[CuL(OH)]\,[H]/[CuL]$	k_1	−7·90
$[CuL(OH)_2]\,[H]/[CuL(OH)]$	k_2	−9·35
$[FeLH]\,[H]^2/[Fe]\,[H_3L]$	$^*K_{12}$	1·68; 1·72[c]; 1·70[d]
$[FeLH]/[Fe]\,[HL]$	β_{1H}	10·30
$[FeL]\,[H]/[FeLH]$	$^*K'$	−3·50
$[FeL]/[Fe]\,[L]$	β_1	16·22
$[FeL(OH)]\,[H]/[FeL]$	k_1	−5·70
$[FeL(OH)_2]\,[H]/[FeL(OH)]$	k_2	−9·18

By evaluation of: [a] UV absorption curves; [b] equimolar solutions; [c] the *A*–pH curve for $c_L = 1{\cdot}5 \cdot 10^{-4}$ M; [d] the A–c_L curve for pH 1·49.

in aqueous medium. Average values of the dissociation constants are summarized in Table I.

UV absorption curves of the reagent in acid medium do not change with pH up to pH 7 (λ_{max} 196 nm). At higher pH values the curves change, and a new maximum is formed at 216 nm. This absorption spectrum is common for all aminohydroxamic acids having one —$CH_2CONHOH$ group bound to amino nitrogen. This indicates the fact that the rest of the molecule (besides the hydroxamic group) affects the position of absorption maximum but slightly, and protonation of amino nitrogen resp. carboxyl group is insignificant in this region.

By evaluating the UV absorption curves of the reagent as a dependence $A = f(pH)$ it was found that the change of spectrum lies in the same pH region as the buffer region of the potenciometric titration curve for calculation of dissociation constant relating hydroxamic group. Table I gives the dissociation constant value from spectrophotometric data, and Table II summarizes spectrophotometric characteristics of the reagent.

IR spectra were measured in KBr discs (1 mg of the compound + 400 mg KBr) within 200 to 4000 cm^{-1}. The absorption bands are denoted according to intensity: s (strong), m (medium), w (weak). Dipotassium salt of N,N-bis(carboxymethyl)-aminoacetohydroxamic acid (in cm^{-1}): 3200 m, 1660 m, 1580 s. 1390 s, 1320 m, 1250 w, 1070 w, 1000 s, 970 w, 900 m, 840 w, 810 s, 720 m, 670 s. Vibration of the

TABLE II

Spectrophotometric Characteristics of N,N-Bis(carboxymethyl)aminoacetohydroxamic Acid and its Chelates with Cu(II) and Fe(III)

Compound	pH	λ_{max} nm	ε $cm^2\ mmol^{-1}$
H_3L, H_2L^-, HL^{2-}	<8	196	5 400
L^{3-}	>10	216	6 150
$HL^{2-} \rightleftarrows L^{3-}$ isosb. point		202	5 000
CuLH	<3	770	51·0
CuL^-	4—7	730	73·6
$CuL(OH)^{2-}$	7·5—8·5	690	59·5
$CuL(OH)_2^{3-}$	9—11	620	87·5
$FeLH^+$	<2	280	3 400; 3 460[a]
FeL	2·5—4·5	436	1 230
$FeL(OH)^-$	5—7·5	436	1 670
$FeL(OH)_2^{2-}$	8—10	—	—

[a] From the molar ratios at pH 1·49.

following groups can be assigned to the individual absorption bands: 3200 cm^{-1} (N—H); 1660 cm^{-1} (C=O, amide I); 1580 cm^{-1} (C=O asym, ionis. carboxyl); 1390 cm^{-1} (COO^- sym); 1320 cm^{-1} (COO^- sym); 1250 cm^{-1} (C—N amide III). The other absorption bands are not characteristical for the structure of the compound given, being identical with those of nitrilotriacetic acid given below. Nitrilotriacetic acid[5]: 3050 s, 2850 w, 2400 w, 1730 s, 1450 s, 1370 w, 1320 m, 1240 m, 1200 m, 1000 s, 960 s, 900 s, 850 m, 740 s, 670 w.

Introduction of hydroxamic functional group in the skeleton of nitrilotriacetic acid lowers the basicity of amino nitrogen by more than 3 orders of magnitude, whereas acidity of carboxyl group (pK_{a_1} 2·44) is the same as that of iminodiacetic acid (pK_{a_1} 2·65, ref.[6]) resp. nitrilotriacetic acid (pK_{a_2} 2·49, ref.[7]). From these facts it follows that the studied reagent H_3L is present in solution as a dipolar ion (in analogy with nitrilotriacetic acid).

System Cu(II)– N,N-*Bis(carboxymethyl)aminoacetohydroxamic Acid*

Formation of cupric chelates was followed in equimolar solutions and solutions with excess reagent in the pH range 0·35 to 11·25. The detailed study in broad pH range was enabled by the adaptation[1] described in Experimental. The evaluated *A*–pH. curve (Fig. 1) indicates formation of several reaction products in the whole pH region

The first blue chelate (λ_{max} 770 nm) is formed in acid region being stable in the pH range 2 to 3. All the Job curves of isomolar solutions of pH 2·5 exhibit a maximum within 620 to 770 nm corresponding to the ratio M : L = 1 : 1. On the basis of these

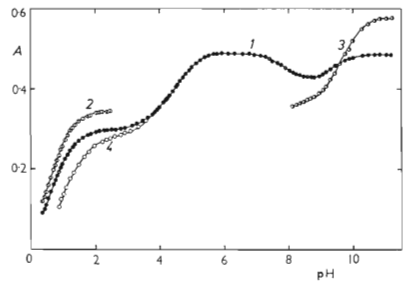


FIG. 1

Curves of Dependence of Absorbance on pH of Solutions of System Cu(II)–N,N-Bis(carboxymethyl)aminoacetohydroxamic Acid

$c_M = 2{\cdot}0 \cdot 10^{-3}$M; $l = 3{\cdot}48$ cm. Curve *1* 700 nm, *2* 770 nm, *3* 620 nm, $c_L = 1{\cdot}0 \cdot 10^{-2}$M; curve *4* 700 nm, $c_L = 2{\cdot}0 \cdot 10^{-3}$M.

findings we analyzed the first ascending section of *A*–pH curves, and graphical analysis according to Eqs (*2*), (*3*) for solutions with excess reagent is linear only for $q = 2$. Also for equimolar solutions the dependence (*5*) is linear only for $q = 2$, the found values of molar absorption coefficient being equal in the both cases (Table II). Number of the split off protons was confirmed by logarithmic analysis according to Eqs (*4*), (*6*), the slope of the straight lines being $\mathrm{tg}\,\alpha = 1{\cdot}9$ to $2{\cdot}05$. Therefrom it follows that in solutions of pH < 3 equilibrium (*D*) is established:

$$Cu^{2+} + H_3L \rightleftharpoons CuLH + 2\,H^+ . \qquad (D)$$

The second gren-blue chelate is formed in the pH region 3·2 to 5·5 and is stable up to pH 7. As also this chelate has the composition of the ratio M : L = 1 : 1 (according to the Job curves of solutions of pH 6·0), it must be supposed that, in the pH region 3·2 to 5·5, dissociation of the protonated chelate takes place. This presumption is confirmed by graphical analysis of the second ascending section of the *A*–pH curve; the dependence (*9*) is linear only for $q = 1$ at all wavelengths within 620 to 770 nm. Hence, equilibrium (*E*) is established in the pH region 3·2 to 5·5 forming the "normal" chelate:

$$CuLH \rightleftharpoons CuL^- + H^+ . \qquad (E)$$

Further increase in pH of the solutions (pH > 7) causes the absorption maximum to shift gradually to lower wavelengths, two green reaction products being formed. All the absorption curves of the solutions of pH 7 to 8·8 form an isosbestic point near 650 nm (Fig. 2). Graphical analysis of descending and ascending sections of the

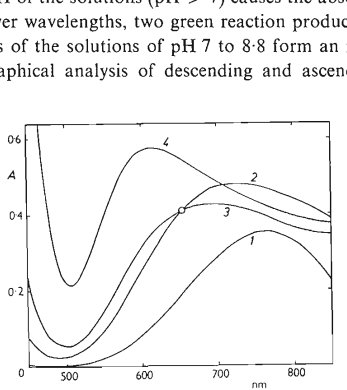


FIG. 2

Absorption Curves of Cupric Chelates of N,N-Bis(carboxymethyl)aminoacetohydroxamic Acid $c_M = 2{\cdot}0 \,.\, 10^{-3}$M, $c_L = 1{\cdot}0 \,.\, 10^{-2}$M, $l = 3{\cdot}48$ cm; curve, pH: *1* 2·17, *2* 6·96, *3* 8·50, *4* 11·0.

A–pH curves by using Eq. (*12*), (*13*) gives straight lines only for $q = 1$ in the both cases. Also the logarithmic dependence (*14*) is linear with $\mathrm{tg}\,\alpha = 0{\cdot}95$ to $1{\cdot}02$, which indicates splitting off of only one proton. Hence in alkaline medium hydroxo complexes are gradually formed:

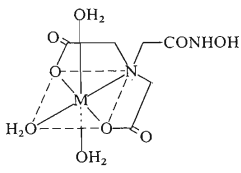
$$\text{pH } 7-8{\cdot}8 \quad \mathrm{CuL^-} + \mathrm{H_2O} \rightleftharpoons \mathrm{CuL(OH)^{2-}} + \mathrm{H^+} \qquad (F)$$

$$\text{pH } 9-11 \quad \mathrm{CuL(OH)^{2-}} + \mathrm{H_2O} \rightleftharpoons \mathrm{CuL(OH)_2^{3-}} + \mathrm{H^+} \qquad (G)$$

Equilibrium constants of all the reactions were calculated from the logarithmic dependences under the condition Y = O (expression on the left side of the equations). The obtained values of the individual constants are given in Table I; spectral characteristics of the chelates are given in Table II.

The study of cupric chelates both in equimolar solutions and in solutions with excess reagent confirmed only the chelates of the composition M : L = 1 : 1. Co-ordination with Cu(II) gives at first the protonated chelate CuLH, as it is the case with other polyfunctional reagents, too. This chelate resembles the chelate CuL of iminodiacetic acid (λ_{max} 750 to 760 nm, $\varepsilon = 76$, $\log \beta_1 = 10{\cdot}3$, ref.[6]) both in optical properties and stability. These facts allow to draw the conclusion that amino nitrogen and the both carboxyl groups take part in co-ordination with Cu(II) in the studied chelate CuLH, the acetohydroxamic group being not involved (*I*).

After splitting off of the proton, hydroxamic group in the chelate $\mathrm{CuL^-}$ co-ordinates in the position above resp. below the planar structure (*II*). This chelate is close to that of nitrilotriacetic acid ($\mathrm{CuL^-}$, ref.[7]) in its properties and stability, and it exhibits a broad absorption band at about 730 nm.



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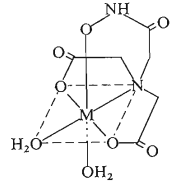

II

In alkaline medium the hydroxo complexes $\mathrm{CuL(OH)^{2-}}$ and $\mathrm{CuL(OH)_2^{3-}}$ are gradually formed containing hydroxyl group instead of water molecule.

System Fe(III)–N,N-*Bis(carboxymethyl)aminoacetohydroxamic Acid*

Solutions of Fe(III) containing a 10 fold excess of the reagent at pH < 2 exhibit no absorption band in visible region. However, complex formation is proved by measurements of the solutions in UV region where a marked absorption band at 280 nm is observed. Detailed measurements at various concentrations again were carried out with the use of the adaptation[1]. Fig. 3 gives one of the obtained *A*–pH dependences.

Composition of the complex, which is formed to the highest degree at pH 1·5, was followed by the method of molar ratios[8]. This titration with the reagent solution at constant pH was carried out again in the 100 ml quartz cell. The evaluated dependence $A = f(c_L/c_M)$ has exponential course; however, graphical analysis using Eqs (*2*), (*3*) gives straight line for $n = 1$ only, which indicates the existence of chelate of the composition M : L = 1 : 1. The value ε obtained by this analysis is close to those obtained by evaluation of *A*–pH curves (Table II).

Graphical analysis of the first ascending section of *A*–pH curve (pH < 2) using Eqs (*2*), (*3*) is linear for $q = 2$ only, being not linear for $q = 1$. Also logarithmic analysis shows that the reaction liberates two protons ($\mathrm{tg}\,\alpha = 1{\cdot}94$ to $2{\cdot}02$). As neither the proton concentration change nor the metal ion–ligand concentration ratio change brings about any change in the position of absorption maximum, the equilibrium (*H*) is established in the solutions under given conditions, only one complex being formed.

$$Fe^{3+} + H_3L \rightleftharpoons FeLH^+ + 2\,H^+ \qquad (H)$$

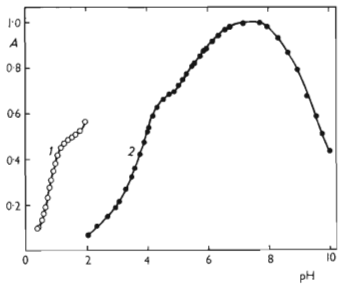

FIG. 3

Curves of Dependence of Absorbance on pH of Solutions of System Fe(III)–N,N-Bis(carboxymethyl)aminoacetohydroxamic Acid

$c_M = 1{\cdot}5 \,.\, 10^{-4}$M, $c_L = 1{\cdot}5 \,.\, 10^{-3}$M. Curve *1* 280 nm, *l* 1 cm; *2* 436 nm, *l* 4 cm.

In solutions of pH > 2 a wine-red complex is formed, all absorption curves of the solutions up to pH 8 exhibiting one absorption band with the maximum at 436 nm. In the pH region 2 to 7 the equilibrium is established more slowly; therefore, the solutions were measured 120 minutes after their preparation. Evaluation of A–pH curve (Fig. 3) shows formation of two complexes in this region, the individual ascending sections of A–pH curves being not markedly separated by horizontal section. The method of molar ratios proved that the complexes existing at pH 5 and at pH 7 again have the composition ratio M : L = 1 : 1.

The graphical analysis of the A–pH curve in the pH range 2·5 to 5 using Eqs (*8*), (*9*) is linear for $q = 1$ only, and logarithmical dependence (*10*) also gives the straight lines indicating unambiguously liberation of one proton. Thus it must be presumed that, in the mentioned region, dissociation of the protonated chelate takes place according to equilibrium (*J*):

$$FeLH^+ \rightleftharpoons FeL + H^+ . \qquad (J)$$

This chelate, however, is stable only in a narrow pH range, being hydrolyzed in solutions of pH > 5. Analysis of this part of A–pH curve using Eqs (*12*) to (*14*) confirms that one proton is liberated in the reaction. As again the complex has the composition M : L = 1 : 1, the equilibrium (K) is established in solutions of pH 5 to 7 to give a hydroxo complex:

$$FeL + H_2O \rightleftharpoons FeL(OH)^- + H^+ . \qquad (K)$$

In solutions of pH 8 to 10 a further hydroxo complex is formed:

$$FeL(OH)^- + H_2O \rightleftharpoons FeL(OH)_2^{2-} + H^+ . \qquad (L)$$

The individual equilibrium constants evaluated from the logarithmic dependences are given in Table I, spectral characteristics of the chelates are given in Table II.

The reagent studied gives with Fe(III) (in analogy with Cu(II)) only water-soluble complexes of the composition M : L = 1 : 1 in the whole pH range studied. The colourless chelate $FeLH^+$ existing in solutions of pH < 2 is similar in its properties to the chelate FeL^+ of iminodiacetic acid (λ_{max} 280 nm; log β_1 = 10·72; ref.[9]). It is obvious that in this chelate Fe(III) is co-ordinated only with amino nitrogen and both carboxyl groups to give two five-membered chelate rings (*I*). First in the chelate FeL formed in the pH region 2 to 5 the hydroxamic group is involved in co-ordination, too, and the chelate formed is wine-red (λ_{max} 436 nm). From these results it follows unambiguously that the hydroxamic group of the studied reagent (and of other aminohydroxamic acids, in contrast to aliphatic and aromatic hydroxamic acids) acts as one-donor only and co-ordinates through oxygen atom to form a six-mem-

bered chelate ring (*II*). The stability constant of the "normal" chelate of the studied reagent is the same as that of nitrilotriacetic acid (Fe-NTA log $\beta_1 = 15{\cdot}9$, ref.[7]). However, this chelate is not stable in a broad pH range, being hydrolyzed to hydroxo and dihydroxo chelates (as the chelate from nitrilotriacetic acid).

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