J. Chem. Eng. Data **1996,** *41,* 821−824 **821**

Equilibria of Complex Formation between Bivalent Metal Ions and 3,3′-Dimethyl-5,5′-bis[(*N*,*N*-bis(carboxymethyl)amino)methyl]-phenolphthalein

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Interactions of 3,3′-dimethyl-5,5′-bis[*N*,*N*-bis(carboxymethyl)amino)methyl]phenolphthalein complexon (OCP) with some of the divalent metal ions (viz., Mn(II), Co(II), Cu(II), Zn(II), Pb(II), and Cd(II)) have been studied by potentiometry in aqueous medium at ionic strength (I) = 0.1 mol dm^{-3} $NaClO_4$ and t = (30 ± 0.1) °C. These metal ions form 1:1 and 2:1 (metal:OCP) chelates with OCP. A study over a wide pH suggests the formation of various stable complex species (viz., MH_2L^{2-}, MHL^{3-}, and ML^{4-} dominant species occurred for a 1:1 metal:ligand ratio and MH_2L^{2-} and M_2L^{2-} species occurred for a 2:1 metal: ligand ratio). The equilibrium constants along with the percentage of metal ions distributed among various complex species formed are given. The proton−ligand dissociation constants for OCP have also been evaluated.

Introduction

3,3′-Dimethyl-5,5′-bis[(*N*,*N*-bis(carboxymethyl)amino)methyl]phenolphthalein complexon (OCP, H_6L) having six dissociable hydrogen atoms and two amino carboxylic groups on each side of its large phthalein nucleus is widely employed for the determination of various alkali and alkaline earth ions present in various biologically important materials. The aqueous solution of the reagent is colorless below pH = 8 and becomes pink above pH = 8.

Kotoucek et al. (1976) studied the interaction of OCP with Ni(II) ions in solution using spectrophotometric methods. They found at pH = 10.55, a 1:1 metal−dye chelate formed with the stability constant value of 7.58 × , further at pH 11 a 2:1 metal−dye stoichiometry was found when there was an excess of Ni(II) ions. V(V) forms a stable complex at pH 2.5 to 3.5 in the presence of fluoride ions in water−acetone (Smirnov et al., 1976). They found that OCP was suitable for precise determination of V(V) ions in spectrophotometric analyses at 610 nm. Abo-El-Wafa et al. (1985) studied the complexation behavior of OCP with rare earth and transition metal cations by employing the spectrophotometric method. Solid complexes of OCP with transition metals (Ni(II), Co(II), Cu(II)) were isolated by Abd-Elmottaleb et al. (1990), having 1:1 [M(OCP)]·$2H_2O$ and 2:1 [M_2(OCP)]·$4H_2O$ stoichiometries.

Because of the suitability of the triphenylmethane group of dyes for metal ion determinations, Srivastava et al. (1995) studied the interaction of these dyes with transition metal ions and determined their stability constant values by potentiometric methods. This paper deals with the determination of stability constants for the divalent metal ions Mn(II), Co(II), Cu(II), Zn(II), Cd(II), and Pb(II) with OCP at t = (30 ± 0.1) °C and ionic strength (I) = 0.1 mol dm^{-3} $NaClO_4$ using potentiometric methods.

Experimental Section

Procedure. The potentiometric measurements over the entire pH range were made with a Beckman OTM 71 pH meter with a gel-filled combination electrode. The titration was carried out in a 100 cm^3 jacketed thermostated titration cell, and the inert atmosphere of nitrogen gas was created by bubbling oxygen free nitrogen through the test solution in the titrating vessel. The following four mixtures were prepared separately and titrated against a standard 5.0 × 10^{-1} mol dm^{-3} sodium hydroxide solution: (a) 5 cm^3 of 0.1 mol dm^{-3} perchloric acid, (b) 5 cm^3 of 0.1 mol dm^{-3} perchloric acid + 5 cm^3 of 1.0 × 10^{-3} mol dm^{-3} OCP solution, (c) 5 cm^3 of 0.1 mol dm^{-3} perchloric acid + 5 cm^3 of 1.0 × 10^{-3} mol dm^{-3} OCP solution + 5 cm^3 of 1.0 × 10^{-3} mol dm^{-3} metal ion solution, and (d) 5 cm^3 of 0.1 mol dm^{-3} perchloric acid + 5 cm^3 of 1.0 × 10^{-3} mol dm^{-3} OCP solution + 10 cm^3 of 1.0 × 10^{-3} mol dm^{-3} metal ion solution. Before the titration was performed, the ionic strength of each set was maintained at 0.1 mol dm^{-3} by addition of an appropriate volume of 1 mol dm^{-3} sodium perchlorate solution. Also the initial volume of each set was kept at 50 cm^3 by addition of double-distilled water. The pH measurements were made at t = (30 ± 0.1) °C using a Julabo (F-10) thermostat. log K_w for all the systems defined in terms of log([H^+][OH^-]) was found to be −13.88 at I = 0.1 mol dm^{-3}.

Materials. Sodium perchlorate, perchloric acid (Merck extrapure product), and sodium hydroxide (AR grade) were used to prepare the solutions in double-distilled water. The solutions were standardized as described by Vogel (1970).

All the bivalent metal salt (Merck extrapure products) solutions were prepared with double-distilled water and were standardized complexometrically by EDTA titrations using appropriate indicators (Flaschke, 1964).

A stock solution (1.0 × 10^{-2} mol dm^{-3}) of OCP (AR grade) was prepared by mass using double-distilled water containing a few drops of 1 mol dm^{-3} NaOH solution. A clear reddish solution was obtained which was diluted further to the experimental concentrations.

The double-distilled water was obtained from the deionized water redistilled over alkaline permanganate with the result being boiled to expel carbon dioxide and cooled down in well-stoppered Pyrex flasks. The pH of this water was 6.8.

Results and Discussion

Proton−Ligand Dissociation Constants. Plots have been made of pH against the moles of alkali required per

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S0021-9568(95)00307-4 CCC: $12.00

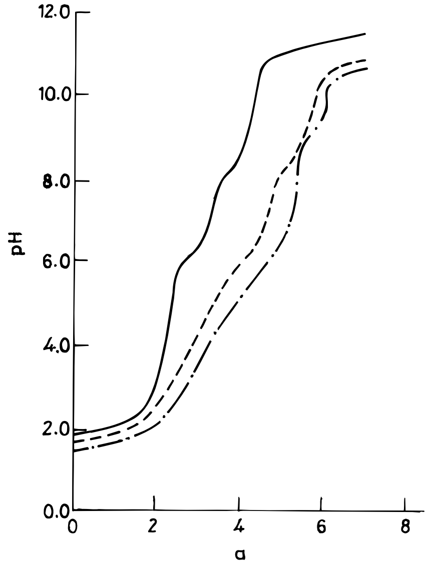

Figure 1. Potentiometric titration curves of Cu^{II}OCP: (—) OCP; (- - -) 1:1 Cu(II):OCP; (—•—) 2:1 Cu(II):OCP.

Table 1. Equilibrium Constants (+3 Standard Deviation) for the Formation of Various Complex Species in 1:1 Bivalent Metal:OCP Mixtures at t = ± 0.1) °C and I = 0.1 mol dm^{-3} NaClO$_4$^{a}

metal ion	$\log_{10} K$ 1	2	3	4	
Mn(II)	5.68 ± 0.04	7.88 ± 0.22	10.71 ± 0.88	11.59 ± 1.02	i
		7.88 ± 0.21	10.72 ± 0.86	11.60 ± 1.00	ii
Pb(II)	7.75 ± 0.02	8.10 ± 0.34	10.85 ± 0.76	11.95 ± 1.22	i
		8.14 ± 0.34	10.87 ± 0.70	11.93 ± 1.20	ii
Cd(II)	6.20 ± 0.03	8.63 ± 0.40	12.21 ± 0.74	13.59 ± 1.14	i
		8.63 ± 0.41	12.22 ± 0.70	13.59 ± 1.16	ii
Co(II)	6.35 ± 0.02	9.44 ± 0.38	12.90 ± 0.82	14.13 ± 1.06	i
		9.44 ± 0.40	12.91 ± 0.86	14.13 ± 1.06	ii
Zn(II)	7.22 ± 0.01	9.10 ± 0.06	12.69 ± 0.96	14.37 ± 0.98	i
		9.10 ± 0.18	12.69 ± 0.96	14.37 ± 0.90	ii
Cu(II)	7.96 ± 0.04	9.78 ± 0.20	13.97 ± 0.48	16.40 ± 1.08	i
		9.80 ± 0.18	13.96 ± 0.50	16.40 ± 1.10	ii

Reactions: (1) $M^{2+} + OH^- \rightleftharpoons M(OH)^+$: (2) $M^{2+} + H_2L^{4-} \rightleftharpoons L^{2-}$; (3) $M^{2+} + HL^{5-} \rightleftharpoons MHL^{3-}$; (4) $M^{2+} + L^{6-} \rightleftharpoons ML^{4-}$. Key to column 6: (i) values obtained by taking the metal hydroxo species into account; (ii) values obtained by ignoring metal hydroxo species.

mole of ligand (a) (e.g. Figure 1), which shows inflections = 2, 4, and 5, and the a values increases to 6. This indicates the dissociation of six protons from OCP. The dissociation constant values of OCP were evaluated using the method described by Nayan and Dey (1976). The values of K_a^1, K_a^2, K_a^3, K_a^4, K_a^5, and K_a^6 were found to be $^{2.16}$, $10^{-3.18}$, $10^{-6.80}$, $10^{-7.96}$, $10^{-10.88}$, and $10^{-11.20}$. These values are quite in line with literature values (Anderegg et al., 1954) obtained by spectrophotometric methods.

The addition of alkali to a metal ion solution causes hydrolysis and forms the hydroxo species in the solution (Nayan and Dey, 1976). The equilibrium involved is

$$M^{2+} + OH^- \rightleftharpoons M(OH)^+ \qquad K = \frac{[M(OH)^+]}{[M^{2+}][OH^-]} \qquad (1)$$

The first base hydrolysis constants (K) for the divalent metal ions have been calculated and are given in Table 1.

One to One Metal–OCP Equilibria. The plot of pH against a for these metal ions with OCP (Figure 1) indicates three dominant inflections at a = 4, 5, and 6, indicating the formation of three stable 1:1 complex species, i.e., MH_2L^{2-}, MHL^{3-}, and ML^{4-}, respectively. Mathematical equations have been derived to determine the various metal–ligand equilibria and equilibrium constants. The equations are based on the papers by Martell et al. (1989) and Nayan (1980).

Table 2. Proton Dissociation Constants (+3 Standard Deviation) for Bivalent Metal-OCP Chelates at t = (30 ± 0.1) °C and I = 0.1 mol dm^{-3} NaClO$_4$^{a}

metal ion	$-\log_{10} K$ 1	2	
Mn(II)	8.04 ± 0.88	10.32 ± 1.22	i
	8.04 ± 0.82	10.32 ± 1.20	ii
	8.20 + 0.92	10.40 ± 1.24	iii
Pb(II)	8.17 ± 0.94	10.12 ± 1.20	i
	8.14 ± 0.90	10.10 ± 1.21	ii
	8.20 ± 0.92	10.34 ± 1.32	iii
Cd(II)	7.29 ± 0.76	9.82 ± 1.42	i
	7.29 ± 0.76	9.82 ± 1.32	ii
	7.32 ± 0.70	10.00 ± 1.38	iii
Co(II)	7.41 ± 0.88	9.97 ± 1.20	i
	7.41 ± 0.82	9.97 ± 1.25	ii
	7.50 ± 0.98	10.02 ± 1.05	iii
Zn(II)	7.29 ± 0.82	9.51 ± 1.08	i
	7.28 ± 0.96	9.49 ± 1.05	ii
	7.42 ± 0.94	9.98 ± 1.00	iii
Cu(II)	6.69 ± 0.95	8.75 ± 0.98	i
	6.70 ± 0.81	8.76 ± 0.96	ii
	6.78 ± 0.95	8.90 ± 0.97	iii

^{a} Reactions: (1) $MH_2L^{2-} \rightleftharpoons MHL^{3-} + H^+$; (2) $MHL^{3-} \rightleftharpoons ML^{4-} + H^+$. Key to column 4: (i) values obtained by taking the metal hydroxo species into account; (ii) values obtained by ignoring the metal hydroxo species; (iii) values obtained by assuming 100% metal ligand association.

Below pH ~ 6.6 an inflection was observed at a = 4, indicating the formation of MH_2L^{2-} complex species, and the equilibrium involved in this region may be expressed as

$$M^{2+} + H_2L^{4-} \rightleftharpoons MH_2L^{2-} \qquad K = \frac{[MH_2L^{2-}]}{[M^{2+}][H_2L^{4-}]} \qquad (2)$$

Again between a = 4 and 5 (pH ≥ 8.2) another inflection occurred, suggesting the formation of the MHL^{3-} species. The equilibrium and equilibrium constant may be represented as

$$M^{2+} + HL^{5-} \rightleftharpoons MHL^{3-} \qquad K = \frac{[MHL^{3-}]}{[M^{2+}][HL^{5-}]} \qquad (3)$$

In this buffer region MH_2L^{2-} is also expected to dissociate into the MHL^{3-} species as

$$MH_2L^{2-} \rightleftharpoons MHL^{3-} + H^+ \qquad K_a^{MH_2L} = \frac{[MHL^{3-}][H^+]}{[MH_2L^{2-}]} \qquad (4)$$

Finally, an inflection is observed at a = 5 and 6, indicating the formation of nonprotonated ML^{4-} species. The equilibria and equilibrium constants involved in this region may be represented by

$$M^{2+} + L^{6-} \rightleftharpoons ML^{4-} \qquad K = \frac{[ML^{4-}]}{[M^{2+}][L^{6-}]} \qquad (5)$$

$$MHL^{3-} \rightleftharpoons ML^{4-} + H^+ \qquad K_a^{MHL} = \frac{[ML^{4-}][H^+]}{[MHL^{3-}]} \qquad (6)$$

The above equilibrium constant values for equations 2–6 have been evaluated for each metal ion and listed in Tables 1 and 2.

Two to One Metal–OCP Equilibria. In order to ascertain the possibility of formation of dinuclear complexes in the solution, a study was extended to a 2:1 initial metal: ligand ratio. For these systems two major inflections were

Table 3. Equilibrium Constants (±3 Standard Deviation) for 2:1 Metal:OCP Ratio at t (30 ± 0.1) °C and $I = 0.1$ mol dm^{-3} $NaClO_4$

	$\log_{10} K$				
metal ion	1	2	3	4[a]	5[a]
Mn(II)	−10.78 ± 0.22	−8.40 ± 0.36	−11.89 ± 0.72	9.31 ± 0.98	15.90 ± 0.96
Pb(II)	−10.44 ± 0.22	−8.70 ± 0.92	−11.44 ± 0.81	9.65 ± 0.96	19.29 ± 0.80
Cd(II)	−9.77 ± 0.38	−8.40 ± 0.88	−11.68 ± 0.72	10.32 ± 0.64	20.72 ± 1.02
Co(II)	−9.54 ± 0.30	−7.80 ± 0.76	−10.80 ± 0.78	10.55 ± 0.85	21.83 ± 1.25
Zn(II)	−10.09 ± 0.22	−6.45 ± 0.44	−10.32 ± 0.60	10.00 ± 0.65	21.76 ± 1.02
Cu(II)	−9.11 ± 0.36	−7.40 ± 0.32	−9.07 ± 0.68	10.98 ± 0.99	23.99 ± 1.02

[a] Measured constants derived from experimental data. Reactions: (1) M^{2+} $H_6I \rightleftharpoons MH_2L^{2-} + 4H^+$; (2) $MH_2L^{2-} \rightleftharpoons MHL^{3-} + H^+$; (3) $M^{2+} + MH_2L^{2-} \rightleftharpoons M_2L^{2-} + 2H^+$; (4) $K_{MH_2L} = K_1/K_a^1K_a^2K_a^3K_a^4$, (5) $K_{M_2L} = K_1K_3/K_a^1K_a^2K_a^3K_a^4K_a^5K_a^6K$ where $K_1 = [MH_2L^{2-}][H^+]^4/[M^{2+}][H_6L]$ and $K_3 = [M_2L^{2-}][H^+]^2/[M^{2+}][MH_2L^{2-}]$.

observed at $a = 4$ and 6, indicating a two-step complexation. In the first step the MH_2L^{2-} species is formed, and in the second step the formation of dinuclear M_2L^{2-} species predominates. The calculation of equilibrium constants for the equilibria involved was based on previous publications (Zompa et al., 1966; Srivastava et al., 1995).

At a = 0−4. Below pH = 5.6 an inflection was observed = 4, indicating the formation of MH_2L^{2-} species in this pH region. The equilibrium may be represented as follows:

$$+ H_6L \xrightleftharpoons{\text{excess of } M^{2+}} MH_2L^{2-} + 4H^+$$

$$K = \frac{[MH_2L^{2-}][H^+]^4}{[M^{2+}][H_6L]} \quad (7)$$

The formation constant K_{MH_2L} has been calculated with the equation

$$K_{MH_2L} = \frac{[MH_2L^{2-}]}{[M^{2+}][H_2L^{4-}]} \quad (8)$$

$$K_{MH_2L} = \frac{K_7}{K_a^1K_a^2K_a^3K_a^4} \quad (9)$$

where K_7 is the equilibrium constant value obtained from eq 7. By using the values of K_7 and proton dissociation constants of OCP (K_a^1, K_a^2, K_a^3, K_a^4), the value of K_{MH_2L} was calculated and is given in Table 3.

Another inflection is observed at $a = 6$ (for pH > 6), indicating the association of a second metal ion with $_2L^{2-}$. The equilibrium involved in the buffer region may be written as

$$+ MH_2L^{2-} \rightleftharpoons M_2L^{2-} + 2\,H^+$$

$$K = \frac{[M_2L^{2-}][H^+]^2}{[M^{2+}][MH_2L^{2-}]} \quad (10)$$

where K is the equilibrium constant for this reaction. It is quite reasonable to expect that the previously formed species MH_2L^{2-} might undergo some degree of acidic dissociation in this pH region. The first acid dissociation constant MH_2L^{2-} lies somewhere between $10^{-8.8}$ and $10^{-10.98}$ for OCP since the ligand H_2L^{4-} dissociates its fifth H^+ in this region. Hence $K_a^{MH_2L}$ has been estimated as usual at $a = 5$ from the pH vs a curves, ignoring the other species. These values are given in Table 3.

The formation constant for M_2L^{2-} [K_{M_2L}] has been evaluated using the following equation and is given in Table 3.

$$K_{M_2L} = \frac{[M_2L^{2-}]}{[M^{2+}][L^{6-}]} \quad (11)$$

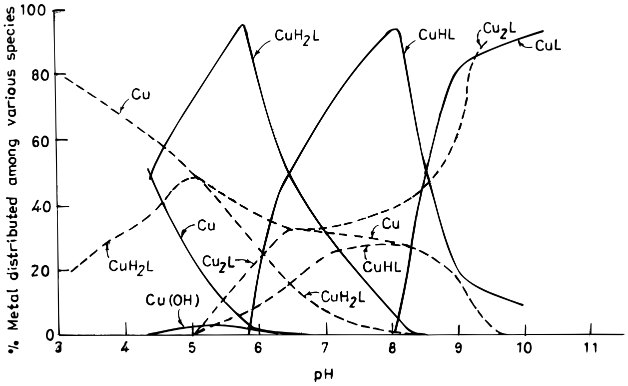

Figure 2. Plots of percentage distribution of Cu(II) against pH: (—) 1:1 Cu(II):OCP; (- - -) 2:1 Cu(II):OCP.

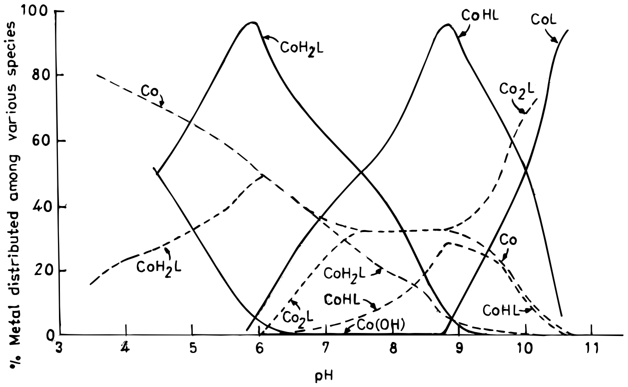

Figure 3. Plots of percentage distribution of Co(II) against pH: (—) 1:1 Co(II):OCP; (- - -) 2:1 Co(II):OCP.

or

$$K_{M_2L} = \frac{K_7K_{10}}{K_a^1K_a^2K_a^3K_a^4K_a^5K_a^6} \quad (12)$$

where K_7 and K_{10} are the equilibrium constants for equilibria (7) and (10), respectively, and K_a^1, K_a^2, K_a^3, K_a^4, K_a^5, and K_a^6 are proton dissociation constants for OCP.

The formation curves (Figure 1) indicate that OCP forms 1:1 as well as 2:1 metal−ligand stoichiometry with these bivalent metal ions, which is in good agreement with earlier reports of Sarkar et al. (1967), Gaizer et al. (1981) for alkaline earth ions, and Murakami et al. (1980) for xylenol orange with divalent metal ions. Moreover Table 1 indicates that no significant change (except in the case of Pb^{2+} and Cu^{2+}) in equilibrium constant values has been observed when it was evaluated by (i) taking into account and (ii) ignoring the simultaneous formation of hydroxo species of the metal ion in the solution. However, in the case of Pb^{2+} and Cu^{2+} a fair amount of deviation is observed, which indicates the simultaneous formation of $Pb(OH)^+$ and $Cu(OH)^+$ species. These results were further confirmed by

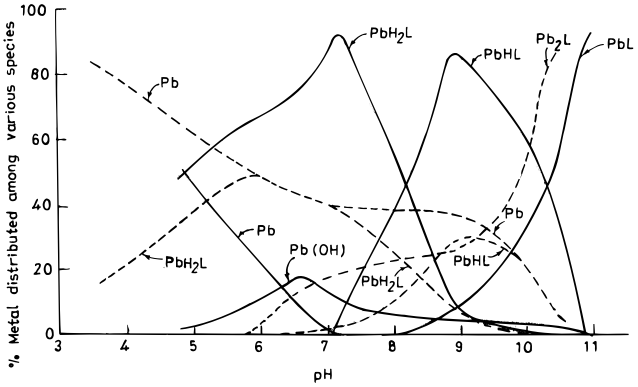

Figure 4. Plots of percentage distribution of Pb(II) against pH: (—) 1:1 Pb(II):OCP; (- - -) 2:1 Pb(II):OCP.

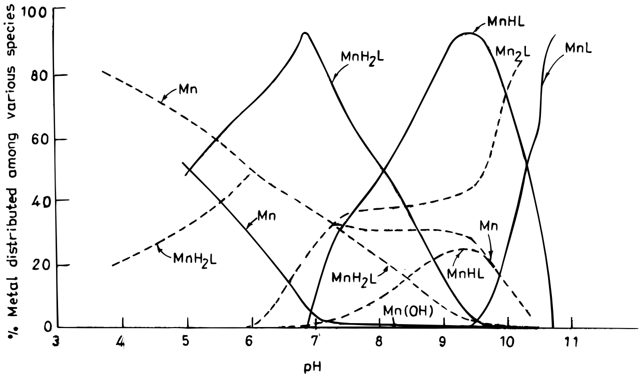

Figure 5. Plots of percentage distribution of Mn(II) against pH:) 1:1 Mn(II):OCP; (- - -) 2:1 Mn(II):OCP.

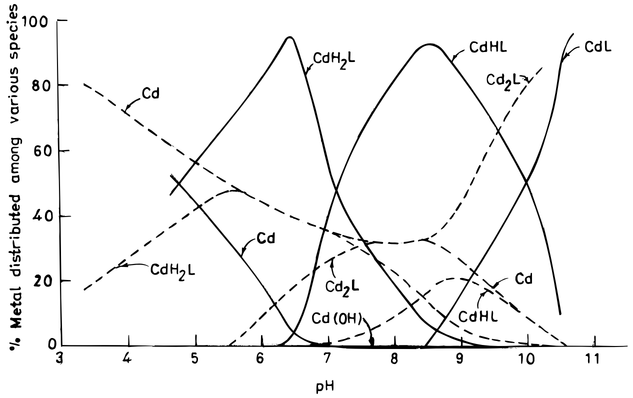

Figure 6. Plots of percentage distribution of Cd(II) against pH:) 1:1 Cd(II):OCP; (- - -) 2:1 Cd(II):OCP.

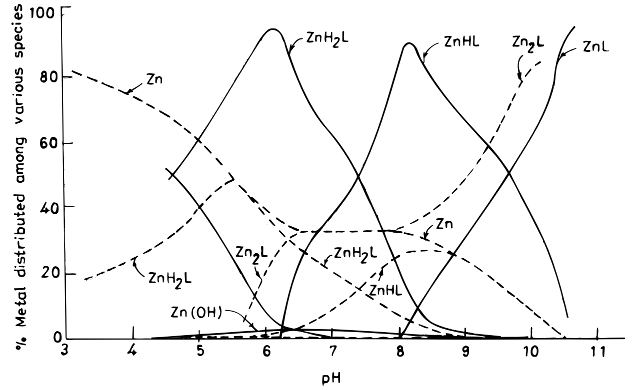

Figure 7. Plots of percentage distribution of Zn(II) against pH: (—) 1:1 Zn(II):OCP; (- - -) 2:1 Zn(II):OCP.

Figures 2−7 (firm line), showing the percent of metal ion distributed as a function of pH. Figure 4 shows that ca. 20% metal simultaneously distributed as its $Pb(OH)^+$ species at the pH ~ 6.5 where the formation of PbH_2L^{2-} species is formed. Similarly, in the case of Cu^{2+} (Figure 2) it forms ca. 5% $Cu(OH)^+$ species at pH ~ 5.6. The stability constant values for 3d divalent ions follow the Irving−William's order, i.e., Mn(II) < Co(II) < Cu(II) > Zn(II) (Irving and William, 1953).

Similarly, Figures 2−7 (dotted lines) reveal the formation of 2:1 metal−ligand stoichiometry by the distribution of percent of metal ions against the entire range of pH. It concludes that at higher pH (10.5−11.5) ca. 95% of the metals distributed in their respective M_2L^{2-} species, whereas in the lower range of pH species MH_2L^{2-} and MHL^{3-} predominate along with the free metal ion.

Acknowledgment

The authors are thankful to the Heads, Departments of Chemistry and Computer Science, Banaras Hindu University, Varanasi, for providing necessary laboratory and computer facilities, respectively.

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Received for review December 7, 1995. Accepted April 16, 1996.⊗

JE950307R

⊗ Abstract published in *Advance ACS Abstracts,* May 15, 1996.