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Complexation Behaviour of Cd(II) with Some Amino Acids and Sulfathiazole at DME

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The present manuscript includes the electro-reduction of Cd(II) ion in the aqueous solution with some amino acids (*e.g.*, DL-threonine, DL-isoleucine, L-serine and L-aspartic acid) and sulfathiazole (STA) at dropping mercury electrode (DME) by treatment of Schaap & McMasters method. All the three types of complexes MXY, MXY_2 and MX_2Y have been found to be reversible and diffusion controlled, involving two electrons in each case (X = amino acid, Y = sulfathiazole). With the help of stability constants of these complexes statistical and electrostatic effects have been considered.

Key Words: Electroreduction, Sulfathiazole, Amino acids, Schaap & McMasters and Stability constants.

INTRODUCTION

Among all sulfadrugs sulfathiazole [4-amino-N-2-thiazolylbeneno-sulphonamide] is clinically one of the most used[1] chemotherapeutic agent employed for the prevention and cure of bacterial infection in humans[1-3]. Metal complexes of sulfathiazole posses many applications as antiviral[4], antibacterial[5], diuretic and antiglaucoma. Rizzotto *et al.*[6] studied the interaction between Hg(II) and sulfathiazole spectroscopically. On the other hand amino acids form biological active complexes, which are important in analytical, biochemical and pharmaceutical field[7-9]. Amino acids forms stable complexes and have analytical importance in separation of transition metals and rare earths[10]. Binary complexes of Cd(II) ion with amino acids have been investigated by Shah *et al.*[11] while complexes of Cd(II) with sulfathiazole have been reported by Narayan *et al.*[12]. Mixed ligand complexes of Cd(II) ion with amino acids and malonic acid have been reported by Sachan *et al.*[13]. Singh *et al.*[14] investigated the complexation behaviour of Cd(II) ion with amino acids and DL-DOPA. Verma *et al.*[15] studied the complexation behaviour of cadmium ion with methyl iminodiacetic acid (MIDA) and some amino acids at DME. The studies of ternary complexes of different metal ions with different ligands have been carried out in our laboratory[16-20]. However, there is no report is available in literature about the mixed ligand complexes of Cd(II) ion with amino acids (such as DL-threonine, DL-isoleucine, L-serine and L-aspartic acid) and sulfathiazole. Hence, the present work has been undertaken for the studies.

EXPERIMENTAL

All the chemicals, sulfathiazole, DL-threonine, DL-isoleucine, L-serine and L-aspartic acid used were of AR grade and their solutions were prepared in double distilled water. Potassium nitrate was used as supporting electrolyte to maintain the ionic strength at 0.2 M. The temperature was maintained constant at 303 ± 2 K. Triton-X-100 of 0.001 % in the final solution have been used as maxima suppresser. The experimental techniques were same as described earlier[21].

RESULTS AND DISCUSSION

Simple systems: Before the study of the mixed ligand complexes, the formation constants of the complexes of Cd with sulfathiazole and Cd with amino acids (DL-threonine, DL-isoleucine, L-serine and L-aspartic acid) were determined by the method of Deford and Hume[22]. The values of formation constants of simple systems are presented in Table-1. The half wave potential of Cd(II) for each series was ranged between -0.580 and -0.583 volt v/s SCE.

TABLE-1
STABILITY CONSTANTS FOR SIMPLE SYSTEMS

Systems	log β_1	log β_2	log β_3
Cd-Sulfathiazole	2.30	4.00	5.26
Cd-DL-Threonine	4.06	7.06	9.02
Cd-DL-Isoleucine	3.90	6.80	8.94
Cd-L-Serine	4.10	7.06	9.08
Cd-L-Aspartic acid	4.40	8.00	10.24

Mixed systems: In case of mixed systems a ligand displacement technique has been used in which the more complexing species is added to a mixture of a metal ion with weaker complexing species. The concentration of weaker ligand (sulfathiazole) was kept constant at two concentrations (0.0002 and 0.002 M) while varying the concentration of strong ligand (amino acids) over a wide range.

In each case, a single well-defined wave was obtained. The plots of E_{de} v/s log I/ I_d–I were linear with a slope of 30 ± 2 mV, showing that the two electrons reduction was reversible. The direct proportionality of the diffusion current to the mercury column indicated that the reduction was entirely diffusion controlled.

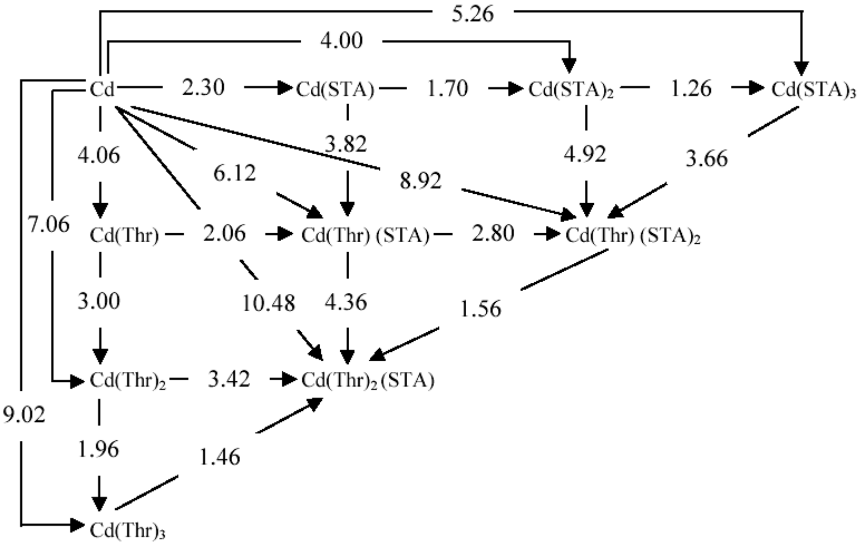
The extended method of Schaap and McMasters[23] was applied to calculate the F_{ij} functions and Leden's[24] graphical extraplotation method to calculate the values of A, B, C and D at the two concentrations of sulfathiazole for each system. Details of calculation are tabulated in Table 3A-6B.

The stability constants β_{11} and β_{12} were evaluated from the two values of B. From the values of C two values of β_{21} were obtained which are in good agreement with each other. The results are presented in Table-2.

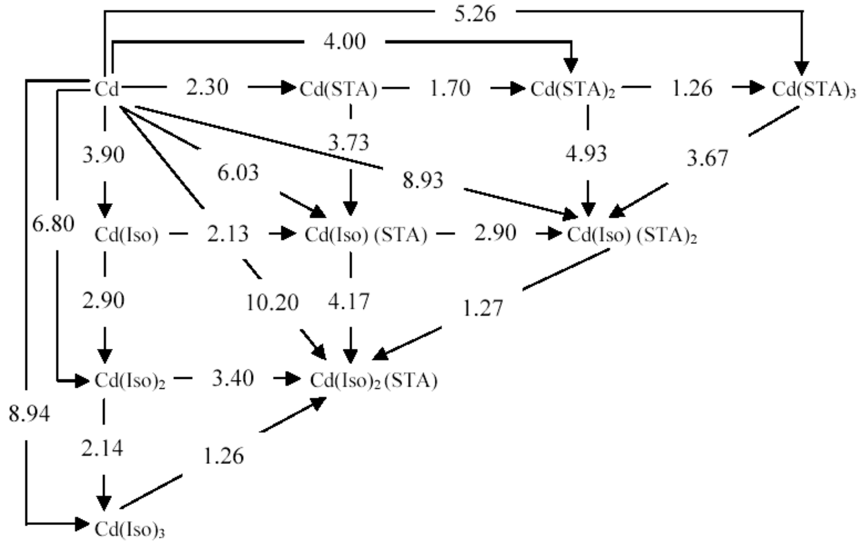
TABLE-2
FORMATION CONSTANTS OF MIXED SYSTEMS

Systems	log β_{11}	log β_{12}	log β_{21}
Cd-DL-Threonine-sulfathiazole	6.12	8.92	10.48
Cd-DL-Isoleucine-sulfathiazole	6.03	8.93	10.20
Cd-L-Serine-sulfathiazole	5.79	9.28	10.55
Cd-L-Aspartic acid-sulfathiazole	6.63	9.52	11.46

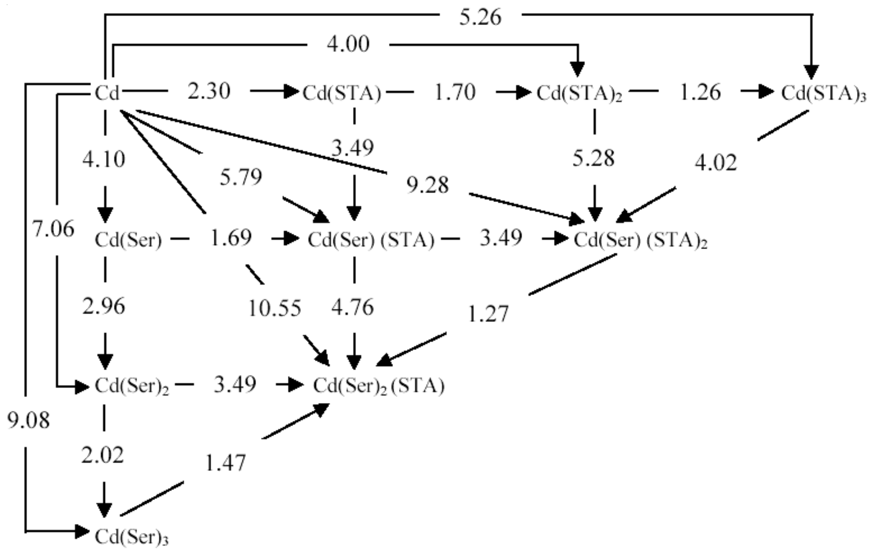
Formation of mixed ligand complexes can be explained by considering the **Schemes I-IV**. The tendency to add [X] (X = amino acids) to Cd[X] and Cd[Y] (Y = sulfathiazole) can be compared. The logarithm value of stability constant of the above complexes are (3.00, 3.82), (2.90, 2.73), (2.96, 3.49) and (3.60, 4.33) for Cd-DL-threonine-STA, Cd-DL-isoleucine-STA, Cd-L-serine-STA and Cd-L-aspartic acid-STA systems, respectively. It indicates the formation of charged complexes.



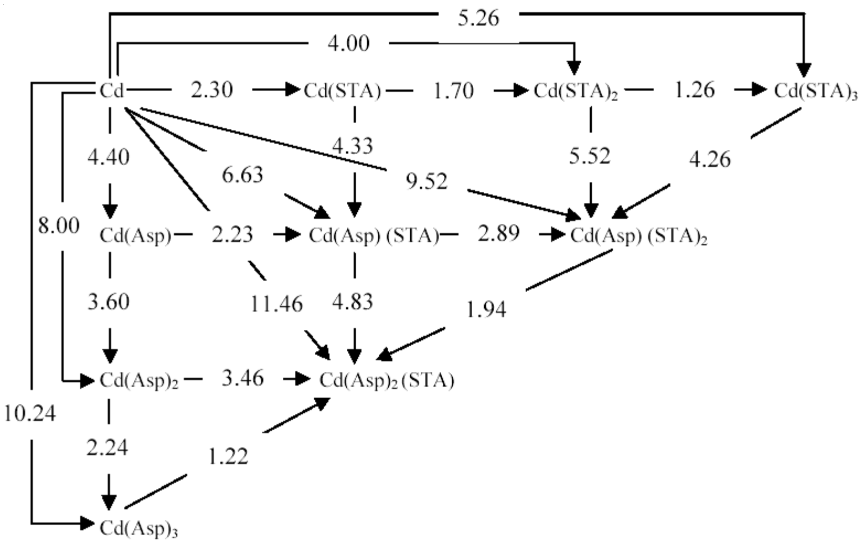
Scheme-I Cd-DL-Threonine-sulfathiazole system



Scheme-II Cd-DL-Isoleucine-sulfathiazole system



Scheme-III Cd-L-Serine-sulfathiazole system



Scheme-IV Cd-DL-Aspartic acid-sulfathiazole system

The tendency to add [Y] to Cd[X] and Cd[Y] can also be compared. The log K values are (2.06, 1.70), (2.13, 1.70), (1.69, 1.70) and (2.23, 1.70) for Cd-DL-threonine-STA, Cd-DL-isoleucine-STA, Cd-L-serine-STA and Cd-L-aspartic acid-STA systems, respectively. This indicates that the addition of sulfathiazole is preferred to Cd[amino acids] as compared to Cd[STA] except the Cd-L-serine system due to some statistical reasons.

The log K values for the addition of [X] to $Cd[X]_2$, Cd[XY] and $Cd[Y]_2$ are (1.96, 4.36, 4.92), (2.14, 4.17, 4.93), (2.02, 4.76, 5.28) and (2.24, 4.83, 5.52) for Cd-DL-threonine-STA, Cd-DL-isoleucine-STA, Cd-L-serine-STA and Cd-L-aspartic acid-STA systems, respectively. This shows that the addition of amino acid is preferred to $Cd[Y]_2$ over $Cd[X]_2$.

The log K values for the addition of [Y] to $Cd[Y]_2$, Cd[XY] and $Cd[X]_2$ are (1.26, 2.80, 3.42), (1.26, 2.90, 3.40), (1.26, 3.49, 3.49) and (1.26, 2.89, 3.46) for Cd-DL-threonine-STA, Cd-DL-isoleucine-STA, Cd-L-serine-STA and Cd-L-aspartic acid-STA systems, respectively. It indicates that addition of sulfathiazole is preferred to $Cd[X]_2$ over $Cd[Y]_2$.

The higher value of β_{21} and β_{30} indicates that ternary complexes are more stable than the binary complexes.

The tendency of formation of the simple and mixed ligand complexes can be expressed by calculating the disproportionation constant K_D for the equilibrium.

$$2Cd[XY] \rightleftharpoons Cd[X]_2 + Cd[Y]_2$$

TABLE-3A

RESULTS FOR Cd-DL-THREONINE-SULFATHIAZOLE SYSTEM

[Sulfathiazole] = 0.0002 M; $E_{1/2}$ [Cd] = 0.580 volt *vs.* SCE

[X] × 10^4	Δ $E_{1/2}$	log I_m/I_c	F_{00}	$F_{10} \times 10^{-3}$	$F_{20} \times 10^{-6}$	$F_{30} \times 10^{-8}$	F_{00} (Cal)	Δ F_{00} (%)
1	0.011	0.01351	2.396	13.560	17.600	10.00*	2.396	0.00
2	0.017	0.04804	4.108	15.340	17.700	10.00*	4.108	0.00
3	0.022	0.05925	6.183	17.143	17.810	10.33*	6.183	0.00
4	0.026	0.07083	8.627	18.967	17.917	10.42	8.627	0.00
5	0.029	0.09382	11.446	20.812	18.024	10.48	11.446	0.00
6	0.031	0.13435	14.646	22.677	18.128	10.47	14.646	0.00
7	0.034	0.12970	18.234	24.563	18.233	10.46	18.234	0.00
8	0.036	0.14894	22.216	26.470	18.337	10.47	22.216	0.00
9	0.038	0.16060	26.599	28.399	18.443	10.48	26.599	0.00
10	0.040	0.16596	31.388	30.348	18.548	10.48	31.388	0.00

A = 1.04 (cal.); B = 1.18 × 10^4; C = 1.75 × 10^7; D = 10.48 × 10^8

*May be deleted

TABLE-3B

RESULTS FOR Cd-DL-THREONINE-SULFATHIAZOLE SYSTEM

[Sulfathiazole] = 0.002 M; $E_{1/2}$ [Cd] = 0.580 volt *vs.* SCE

[X] × 10^4	Δ $E_{1/2}$	log I_m/I_c	F_{00}	$F_{10} \times 10^{-3}$	$F_{20} \times 10^{-6}$	$F_{30} \times 10^{-8}$	F_{00} (Cal)	Δ F_{00} (%)
1	0.018	0.01498	4.110	26.700	72.000	10.00*	4.110	0.000
2	0.027	0.01679	8.224	33.920	72.100	10.00*	8.224	0.000
3	0.033	0.04162	13.789	41.163	72.210	10.33*	13.789	0.000
4	0.038	0.05403	20.811	48.428	72.320	10.50	20.811	0.000
5	0.042	0.06947	29.296	55.712	72.424	10.48	29.296	0.000
6	0.045	0.09669	39.250	63.017	72.528	10.47	39.250	0.000
7	0.047	0.14114	50.679	70.342	72.632	10.46	50.680	0.002
8	0.050	0.13990	63.592	77.690	72.737	10.47	63.592	0.000
9	0.052	0.16202	77.994	85.060	72.844	10.48	77.993	-0.001
10	0.054	0.17603	93.888	92.448	72.948	10.48	93.888	0.000

A = 1.44 (cal.); B = 1.95 × 10^4; C = 7.19 × 10^7; D = 10.48 × 10^8

*May be deleted

TABLE-4A
RESULTS FOR Cd-DL-ISOLEUCINE-SULFATHIAZOLE SYSTEM
[Sulfathiazole] = 0.0002 M; $E_{1/2}$ [Cd] = 0.580 volt *vs.* SCE

[X] × 10^4	$\Delta E_{1/2}$	log I_m/I_c	F_{00}	$F_{10} \times 10^{-3}$	$F_{20} \times 10^{-6}$	$F_{30} \times 10^{-8}$	F_{00} (Cal)	ΔF_{00} (%)
1	0.008	0.02498	1.955	9.150	9.500	7.00*	1.955	0.00
2	0.013	0.5377	3.064	10.120	9.600	8.50*	3.064	0.00
3	0.017	0.07509	4.372	11.106	9.686	8.53*	4.372	0.00
4	0.020	0.10427	5.884	12.110	9.775	8.63*	5.884	0.00
5	0.023	0.11594	7.606	13.132	9.864	8.68	7.606	0.00
6	0.025	0.14793	9.543	14.172	9.953	8.71	9.543	0.00
7	0.027	0.16985	11.699	15.227	10.038	8.69	11.699	0.00
8	0.029	0.18380	14.081	16.301	10.126	8.70	14.081	0.00
9	0.031	0.19116	16.693	17.392	10.214	8.71	16.693	0.00
10	0.033	0.19303	19.541	18.501	10.301	8.71	19.541	0.00

A = 1.04 (cal.); B = 8.20 × 10^3; C = 9.43 × 10^6; D = 8.71 × 10^8

*May be deleted

TABLE-4B
RESULTS FOR Cd-DL-ISOLEUCINE-SULFATHIAZOLE SYSTEM
[Sulfathiazole] = 0.002 M; $E_{1/2}$ [Cd] = 0.580 volt *vs.* SCE

[X] × 10^4	$\Delta E_{1/2}$	log I_m/I_c	F_{00}	$F_{10} \times 10^{-3}$	$F_{20} \times 10^{-6}$	$F_{30} \times 10^{-8}$	F_{00} (Cal)	ΔF_{00} (%)
1	0.014	0.03555	3.172	17.320	38.200	10.00*	3.172	0.00
2	0.021	0.05491	5.670	21.150	38.250	7.50*	5.670	0.00
3	0.027	0.05314	8.942	25.006	38.353	8.43*	8.942	0.00
4	0.031	0.08230	12.992	28.880	38.450	8.75	12.992	0.00
5	0.035	0.08655	17.824	32.768	38.536	8.72	17.824	0.00
6	0.038	0.10577	23.444	36.673	38.621	8.68	23.444	0.00
7	0.040	0.14426	29.858	40.597	38.710	8.71	29.858	0.00
8	0.042	0.17168	37.070	44.538	38.797	8.71	37.070	0.00
9	0.044	0.19016	45.086	48.496	38.884	8.71	45.086	0.00
10	0.045	0.23453	53.911	52.471	38.971	8.71	53.911	0.00

A = 1.44 (cal.); B = 1.35 × 10^4; C = 3.81 × 10^7; D = 8.71 × 10^8

*May be deleted

TABLE-5A

RESULTS FOR Cd-L-SERINE-SULFATHIAZOLE SYSTEM

[Sulfathiazole] = 0.0002 M; $E_{1/2}$ [Cd] = 0.580 volt *vs.* SCE

$[X] \times 10^4$	$\Delta E_{1/2}$	$\log I_m/I_c$	F_{00}	$F_{10} \times 10^{-3}$	$F_{20} \times 10^{-6}$	$F_{30} \times 10^{-8}$	F_{00} (Cal)	ΔF_{00} (%)
1	0.011	0.03457	2.515	14.750	19.500	10.00^*	2.515	0.00
2	0.017	0.07648	4.386	16.730	19.650	12.50^*	4.386	0.00
3	0.022	0.09140	6.658	18.727	19.757	11.90^*	6.658	0.00
4	0.026	0.10537	9.341	20.752	19.880	12.00	9.341	0.00
5	0.029	0.12999	12.440	22.800	20.000	12.00	12.440	0.00
6	0.032	0.13850	15.964	24.873	20.122	12.03	15.964	0.00
7	0.035	0.13482	19.919	26.970	20.243	12.04	19.919	0.00
8	0.037	0.15483	24.312	29.090	20.362	12.03	24.312	0.00
9	0.039	0.16712	29.151	31.234	20.483	12.03	29.151	0.00
10	0.041	0.17303	34.443	33.403	20.603	12.03	34.443	0.00

A = 1.04 (cal.); B = 1.28×10^4; C = 1.94×10^7; D = 12.03×10^8

*May be deleted

TABLE-5B

RESULTS FOR Cd-L-SERINE-SULFATHIAZOLE SYSTEM

[Sulfathiazole] = 0.002 M; $E_{1/2}$ [Cd] = 0.580 volt *vs.* SCE

$[X] \times 10^4$	$\Delta E_{1/2}$	$\log I_m/I_c$	F_{00}	$F_{10} \times 10^{-3}$	$F_{20} \times 10^{-6}$	$F_{30} \times 10^{-8}$	F_{00} (Cal)	ΔF_{00} (%)
1	0.018	0.04558	4.410	29.700	82.000	10.00^*	4.410	0.000
2	0.027	0.05720	9.026	37.930	82.150	12.50^*	9.026	0.000
3	0.033	0.08658	15.293	46.177	82.257	11.90^*	15.293	0.000
4	0.038	0.10162	23.221	54.452	82.381	12.03	23.221	0.000
5	0.042	0.11873	32.815	62.750	82.500	12.00	32.815	0.000
6	0.045	0.14713	44.084	71.073	82.622	12.03	44.084	0.000
7	0.048	0.15917	57.033	79.419	82.741	12.03	57.034	0.002
8	0.050	0.19185	71.672	87.790	82.862	12.03	71.672	0.000
9	0.052	0.21447	88.006	96.184	82.983	12.03	88.006	0.000
10	0.054	0.22890	106.043	104.603	83.103	12.03	106.043	0.000

A = 1.44 (cal.); B = 2.15×10^4; C = 8.19×10^7: D = 12.03×10^8

*May be deleted

TABLE-6A

RESULTS FOR Cd-L-ASPARTIC ACID-SULFATHIAZOLE SYSTEM

[Sulfathiazole] = 0.0002 M; $E_{1/2}$ [Cd] = 0.580 volt *vs.* SCE

[X] × 10^4	$\Delta E_{1/2}$	log I_m/I_c	F_{00}	$F_{10} \times 10^{-3}$	$F_{20} \times 10^{-6}$	$F_{30} \times 10^{-8}$	F_{00} (Cal)	ΔF_{00} (%)
1	0.021	0.02124	5.247	40.070	15.970	1.70	5.247	0.000
2	0.032	0.03981	12.719	58.395	16.147	1.74	12.719	0.000
3	0.040	0.04137	23.560	75.067	16.322	1.74	23.560	0.000
4	0.045	0.08119	37.874	92.085	16.496	1.74	37.874	0.000
5	0.049	0.11613	55.765	109.450	16.670	1.74	55.765	0.000
6	0.052	0.15835	77.337	127.162	16.844	1.74	77.338	0.001
7	0.056	0.14844	102.698	145.226	17.018	1.74	102.698	0.000
8	0.059	0.15748	131.950	163.638	17.192	1.74	131.949	-0.001
9	0.061	0.18853	165.196	182.396	17.366	1.74	165.195	-0.001
10	0.064	0.17723	202.540	201.500	17.540	1.74	202.540	0.000

A = 1.04 (cal.); B = 2.61 × 10^4; C = 1.58 × 10^8; D = 1.74 × 10^{10}

TABLE-6B

RESULTS FOR Cd-L-ASPARTIC ACID-SULFATHIAZOLE SYSTEM

[Sulfathiazole] = 0.002 M; $E_{1/2}$ [Cd] = 0.580 volt *vs.* SCE

[X] × 10^4	$\Delta E_{1/2}$	log I_m/I_c	F_{00}	$F_{10} \times 10^{-3}$	$F_{20} \times 10^{-6}$	$F_{30} \times 10^{-8}$	F_{00} (Cal)	ΔF_{00} (%)
1	0.032	0.04585	12.897	11.457	67.770	1.70	12.897	0.000
2	0.046	0.04912	37.979	18.270	67.947	1.74	37.979	0.000
3	0.054	0.08872	76.789	25.116	68.121	1.74	76.790	0.001
4	0.060	0.11585	129.434	31.998	68.296	1.74	129.434	0.000
5	0.065	0.12974	196.015	38.915	68.470	1.74	196.015	0.000
6	0.069	0.14628	276.637	45.866	68.646	1.74	276.638	0.001
7	0.072	0.17441	371.407	52.852	68.818	1.74	371.408	0.001
8	0.075	0.18638	480.429	59.874	68.992	1.74	480.429	0.000
9	0.078	0.18584	603.809	66.930	69.166	1.74	603.805	-0.001
10	0.080	0.20859	741.635	74.019	69.339	1.74	741.640	-0.001

A = 1.44 (cal.); B = 4.68 × 10^4; C = 6.76 × 10^8; D = 1.74 × 10^{10}

The values of log K_D is -0.60 statistically but the observed values are found to be -1.19, -1.27, -0.52 and -1.25 for Cd-DL-threonine-STA, Cd-DL-isoleucine-STA, Cd-L-serine-STA and Cd-L-aspartic acid-STA systems, respectively. More negative values of log K_D for each equilibrium account for the stability of mixed ligand complexes.

For comparing the stabilities of simple and mixed ligand complexes, it is convenient to measure the mixing constants.

$$K_m = \frac{\beta_{11}}{\sqrt{\beta_{02} \times \beta_{20}}}$$

The log K_m values are +0.59, +0.63, +0.26 and +0.63 for Cd-DL-threonine-STA, Cd-DL-isoleucine-STA, Cd-L-serine-STA and Cd-L-aspartic acid-STA systems, respectively. The positive values of mixing and stabilization constants show that the ternary complexes are more stable than the binary complexes.

The tendency to form mixed ligand complexes in solution can be expressed quantitatively by other approach. Δ log K which is the result from the subtraction of two constants and must therefore, be a constant. This corresponds to:

$$\Delta \log K = \log K^{MA}_{MAB} - \log K^{M}_{MB}$$

Since more coordination positions are available for bonding of the first ligand [A] to a given multivalent metal ion than for the second ligand [B].

$$\log K^{M}_{MA} > \log K^{MA}_{MA_2}$$

Usually holds *i.e.* one expects to observe negative values for Δ log K. Another probably more satisfactory manner is to determine statistical values for Δ log K. The statistical values for regular octahedron (oh) is 5/12 and $\Delta \log K_{oh} = -0.4$. For a square planer (sp), the value of $\Delta \log K_{sp} = -0.6$ and for the distorted octahedron (do), the statistical values *i.e.* $\Delta \log K_{do}$ = lie between -0.9 to -0.3.

The Δ log K values can be obtained using the following equations:

$$\Delta \log K_{11} = \log \beta_{11} - (\log \beta_{10} + \log \beta_{01})$$

$$\Delta \log K_{12} = \log \beta_{12} - (\log \beta_{10} + \log \beta_{02})$$

$$\Delta \log K_{21} = \log \beta_{21} - (\log \beta_{20} + \log \beta_{01})$$

The observed values of $\Delta \log K_{11}$, $\Delta \log K_{12}$ and $\Delta \log K_{21}$ are (-0.23, +0.86, +1.12), (-0.17, +1.03, +1.10), (-0.60, +1.18, +1.19) and (-0.07, +1.17, +1.15) for Cd-DL-threonine-STA, Cd-DL-isoleucine-STA, Cd-L-serine-STA and Cd-L-aspartic acid-STA systems, respectively. The Δ log K values are higher than statistical value, which again proves that the ternary complexes are more stable than expected statistically.

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