Stability constant of some Metal Ion Complexes of (6-(2-Amino-2-(4-hydroxy phenyl)-acetamido)-3,3⁻-dimethyl-7-oxo-4-thia-1-aza-bicyclo[3,2,0]heptanes-2-carboxylicacid(Amoxicillin)

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Abstract

Measurement of stability constant of some complexes formed by (6-(2-amino-2-(4-hydroxy phenyl)-acetamido)-3,3-dimethyl-7-oxo-4-thia-1-aza-bicyclo[3,2,0] heptanes-2-carboxylic acid (Amoxicillin) with (Cr^{+3}, Co^{+2}, Ni^{+2}, Cu^{+2},Ag^{+1}, and Cd^{+2}) ions, have been performed(Amoxicillin) was found to behave as bidentate ligand with ahardness parameter(α)of (0.46)and asoftness parameter(β)of (1.03) while complexes being formed were of (1:1)ratio.

Keywords: Amoxicillin, Stability constant, Transation Metals

Introduction

Amoxicillin is a member of penicillin's group which is a very important class of β lactamic antibiotics used in the therapy because of ite their specific toxicity towards bacteria[1].Most of the organic drugs used against bacterial and viral infection contain donor atoms (N,O,S) which easily coordinate with metal ions[2,3]. V.G.Alekseev and coworkers [4,5] studied and determined of stability constants of cobalt(II) and aluminum ions $^{(III)}$ with Amoxicillin , Glycine and cephelexin in aqueous solutions by using pH-Metric titration at $20C^0$ with 0.1(KNO_3). And also S.V. Lapshin and coworkers [6] studied the stability constant of copper ions with ampicillinc, Amoxicillin and cephalexin (L)anions in aqueous solution at 20 C^0 with 0.1 (KNO_3) by using pH-potentiometry. Recently synthesis of some new Schiff bases derived from Amoxicillin tri hydrate with Cinnamaldehyde and p-Chlorobenzaldehyde and their complexes with bivalent transition metal ions Co(II), Zn(II), Ni(II), and Mn(II) , the ligand and their metal complexes were characterized on the basis of elemental analysis and micro analytical datas. [7]
In this work we are interested mainly in measuring the stability constants(Kst) of complexes in aqueous solution of Amoxicillin , also to apply pearsons [8] hard-soft , acid-base postutute (HSAB) to explain the behavior of this ligand interms of misonos parameters[9] .

Experimental

Reagents: The ligand (Amoxicillin) was used (B.D.H) analar grade.

Standard solutions of metal nitrate were prepared from analar reagents.

Solution of metal nitrate were made with distilled water and carbonate free allkali solution and were standardized against pure potasium hydrogen phthalate[10].

Apparatus and proccdure: pH-Measurements were carried out with Philips pH-Meter.

The pH-Meter was standardized before each run against buffer solutions of known pH values and was checked at the end of each run.

Results and calculation

The acid dissociation constant of (Amoxicillin) was calculated and the detailed calculation can be seen in Table(1).

In order to determine stability constant of metal complexes, two founctions must be calculated.The concentrations of the free chelating species (L^-)and the degree of formation(n^-) which is defind as the averge of ligand species bound per atom of metal. Concentration of free ligand species was calculated by equation previously used with thorium-glycinate [10] of the form:

$Log[L^-]=(pH-PKa)+ Log\{(L)_T - [KOH]_T\}$……(1)

An expression for the degree of formation(n^-) was used of the form:-

$$n^- = \frac{[L]_T - [\frac{[H^+]}{Ka} + 1][L^-]}{[M]_T} \dots\dots\dots (2)$$

Where $[L]_T$,$[L^-]$,$[M]_T$, are concentrations of total ligands, free ligands and total metal ion, respectively. For the present systems, the reported stability constants, β_1, β_2 and β_3 were computed using the well known J.Bierum summation equation with$[L^-]$ an$[n^-]$ calculated at different pH values from equations (1)and(2) respectively.

$$\left[\sum_{n=0}^{n=max} [n - n^-]\beta_n [L]^{n^-} = 0\right] \dots\dots\dots (3)$$

It can be shown after simple approximation that equation(3) may be written for the present system as:

$$\frac{1}{\beta_3} + (n^- - 1)[L]\frac{\beta_1}{\beta_3} + (n^- - 2)[L]^2\frac{\beta_2}{\beta_3} = (3 - n^-)[L]^3 \ldots\ldots\ldots (4)$$

Where β_1, β_2 and β_3 are the corresponding stability constants equation(4) is of the form:

C+ aX+ bY=Z
Where

$$X = \frac{(n^- - 1)(L)}{n^-}$$
$$Y = \frac{(n^- - 2)(L)^2}{n^-}$$
$$Z = \frac{(3 - n^-)(L)^3}{n^-}$$

All being experimentally determined functions and
$a = \frac{\beta_1}{\beta 2}$, b = $\frac{\beta_2}{\beta_3}$ and c = $\frac{1}{\beta_3}$

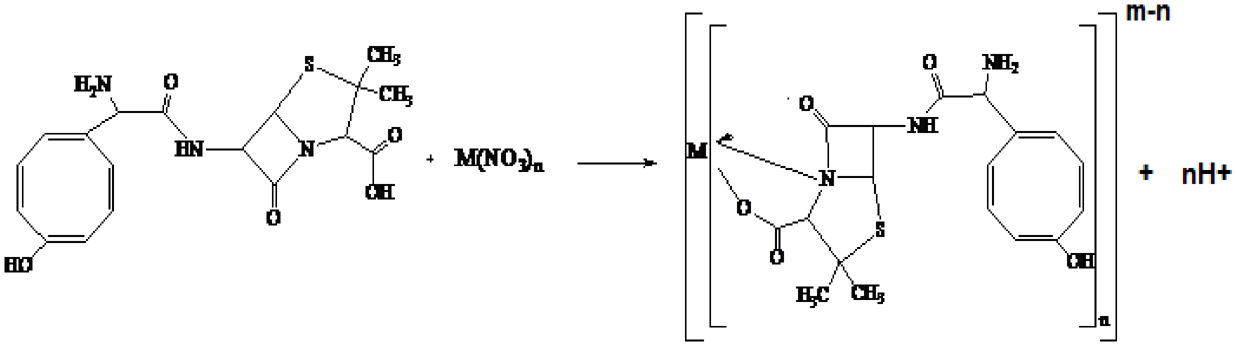
In each system,the functions X,Y and Z were calculated from the experimentel data and where fitted to aggression equation using Cramemer's rule for solving such equation.

Typical titration results are summarized in Tables (2,3,4,5,6,7 and 8).

Stability constants metal- Amoxicillin complexes and Hardness-softness parameter of Amoxicillin are calculated using Misonos equation[8] results which are shown in Table(9).

Discussion

The decrease in the PH value of solutions of (Amoxicillin) when aneutral salt solution of metal ions were added is a clear indication of complexe formation. The chemical equation representing the equllibrium could be written as follows.



However, calculation of acidity as can be seen in Table (1) (Ka=9×10^{-9}) indicates that Amoxicillin behaves as an acid.

It is reasonable to expect acorrelation between the stability of the complex and the acidic dissociation constant of the conjugate acid of the ligand

$$nHL + M^{+m} \rightleftharpoons [ML_n]^{m-n} + nH^+$$

considering the casc of Amoxicillin,the association which metal ions takes the following route: n (Amoxicillin) + M^{+m} ⇌ $[M(Amoxicillin)_n]^{m-n}$ + nH^+
where$[M(Amoxicillin)_n]^{m-n}$ represents

From Table (9) the following conclusion could be drown because of the negative values of (β_2, β_3 , β_4)….etc formation of (1:1) complex species between (Amoxicillin) and metal ions may be considered as the only species existing in solution.

Hardness-Softness parameters

The relationship between stability constant of complexes and the characteristics properties of their constituents was established by Pearson [9] he forwarded the interpretation of hard interaction as eleactrostic and soft one as covalent,

Later, Drago and Wayland put forward a two parametes equation to represent their acid-base interactions[11]misano[8]has introduced aquantitative relationships for coordination compounds which can be expressed as follows

$PK= {}^{-}Log\ K= \alpha X + \beta y + \gamma$…….(5)

Where k is the ((stability constant)) of the complex, X and Y are parameters of metal ion, (α) and (β)those of the ligand .The parameter(γ) is characteristic of Ligand and used to adjust the PKs ,so that all lie on the same scale, .The(Y) parameter is considered to be measure of softness and may be evaluated from atomic parameters including the radius of the ion from atomic parameters including the radius of the ion ,our results for the soft and hard parameters(Y) is complete agreement with those published by misono [8] .The analogous ligand parameter(B) like wise shows the expected increase in values from hard to soft species: OH^{-}=0.4, NH_3=1.08, Cl^{-}=2.4, Br^{-}=5.58,I^{-}=7.17, $S_2O_3^{=}$ =12.4[12]

Softness parameter(B)of(Amoxicillin) Ligand under investigation has value of (1.03) in reminiscent to between of Glycine as ligand [13] (B=1.23)and amine as ligand (B=1.08).

The(α)and(X)parameters were interpreted by Misono as hardness parameters of ligand and Metal ion respectively although it includes the inherent acids-base strength of the ligand and metal ion.

Thus(x)is closely related to the electronegativity of the ion and measures the tendency of metal ion to accept electrons from the ligand. Calculated hardness parameter (α)for ligand (Amoxicillin) was found to be (α =0.46) a value comparable to (0.455)of the (N,N- bis (2-hydroxy ethyl)Glycine[14] the weakly acidic ligand.

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Table No.(1):Determination of the disscociation of Aomoxicine.20ML(0.025M)of Amoxiciline +27.5ml of water temp.:25C^0 , μ =0.1 $HL \rightleftharpoons H^+ + L^-$

$$pKa = pH + Log\,\frac{[HL]+[OH]^-}{[B]-[OH]^-}$$

1	2	3	4	5	6	7
Ml KOH 0.085M	pH	Stoichiometric [B]	[HL]	[OH$^-$]	$\frac{[HL]+[OH]^-}{[B]-[OH]^-}$	pKa=pH+ Logcol 6
0	6.66	0	1.05×10^{-2}	4.5×10^{-8}	0	0
0.5	7.06	8.85×10^{-4}	1.04	1.148×10^{-7}	10.7539	8.09
1	7.35	1.74×10^{-3}	1.03	2.23×10^{-7}	4.922	8.04
1.5	7.57	2.60	1.02	3.71×10^{-7}	2.9345	8.03
2	7.74	3.43	1.01	5.49×10^{-7}	1.8753	8.01
2.5	7.91	4.25	1×10^{-3}	8.12×10^{-7}	1.3534	8.04
3	8.09	5.05	9.9	1.23×10^{-6}	0.9625	8.04
pKa=8.04 , Ka=9×10^{-9}						

Table No.(2):Titration of 30ml(0.025M)of Amoxiciline and 5Ml(0.05M)M(NO$_3$)$_n$ solutions with 0.085M KOH solutions Temp. :25C^0 ,Vol.47.5ml=0.1 ,μ=0.1

Ml KOH (0.085M)	Ag^+	Co^{+2}	Ni^{+2}	Cu^{+2}	Cd^{+2}	Cr^{+3}
0.0	4.38	6.05	5.86	3.31	6.22	4.46
0.5	5.26	6.46	6.13	3.41	6.55	4.56
1.0	6.41	6.7	6.40	3.52	6.75	4.68
1.5	6.85	6.87	6.61	3.68	6.89	4.85
2.0	7.11	6.99	6.74	3.83	7.04	5.04
2.5	7.27	7.08	6.85	4.02	7.16	5.25
3.0	7.41	7.18	6.97	4.24	7.19	5.48
3.5	7.51	7.22	7.08	4.57	7.19	5.61
4.0	7.61	7.32	7.18	4.89	ppt	5.73
4.5	7.73	7.40	7.29	5.19	-	5.83
5.0	7.83	7.44	7.40	5.50	-	5.86

Table No.(3):Titration of (0.025M) Amoxiciline and (0.05M) $Ag(NO_3)$ solution with 0.085M KOH solution.Temp.:25C^0, μ =0.1

Ml KOH 0.085M	pH	$[KOH]_T$	$[M]_T$	$[L]_T$	$\frac{H+}{Ka}$	$[L]^-$	$\bar{n}$
0.0	4.38	0	5.26×10-3	1.57×10^{-4}	4631.88	0	0
0.5	5.26	8.85×10^{-4}	5.21	1.56	610.6	2.44×10^{-5}	0.1274
1.0	6.41	1.74×10^{-3}	5.15	1.54	43.22	3.2×10^{-4}	0.2407
1.5	6.85	2.60	5.10	1.53	15.69	8.19×10^{-4}	0.3165
2.0	7.11	3.43	5.05	1.51	8.62	1.37×10^{-3}	0.3782
2.5	7.27	4.25	5.00	1.50	5.96	1.82×10^{-3}	0.4587
3.0	7.41	5.05	4.95	1.48	4.32	2.28×10^{-3}	0.5334
3.5	7.51	5.83	4.90	1.47	3.43	2.61×10^{-3}	0.6333
4.0	7.61	6.60	4.85	1.47	2.72	2.93×10^{-3}	0.7384
4.5	7.73	7.35	4.81	1.44	2.06	3.45×10^{-3}	0.7970
5.0	7.83	8.09	4.76	1.42	1.64	3.76×10^{-3}	0.8937

1) Calculation of Free Ligand $[L^-]$ from equation
$Log[L^-]=(pH-pKa) + Log[L]_T-[KOH]_T$

2) Calculation of the degree of formation ($\bar{n}$)was used of the from $n^- = \frac{[L]_T-[\frac{[H^+]}{Ka}+1][L^-]}{[M]_T}$

Table No.(4):Titration of (0.025M) Amoxiciline and (0.05M) $Cd(NO_3)_2$ solution with 0.085M KOH solution Temp.:25C^0 ,Vol.47.5ml=0.1 , μ =0.1

Ml KOH 0.085M	pH	$[KOH]_T$	$[M]_T$	$[L]_T$	$\frac{H+}{Ka}$	$[L]^-$	$\bar{n}$
0.0	6.22	0	5.26×10^{-3}	1.57×10^{-4}	66.951	0	0
0.5	6.55	8.85×10^{-4}	5.21	1.56	31.315	4.76×10^{-4}	0.0408
1.0	6.75	1.74×10^{-3}	5.15	1.54	19.758	7	0.1665
1.5	6.89	2.60	5.10	1.53	14.31	8.99	0.3009
2.0	7.04	3.43	5.05	1.51	10.133	1.167×10^{-3}	0.41738
2.5	7.16	4.25	5.00	1.50	7.68	1.41	0.5398
3.0	7.19	5.05	4.95	1.48	7.17	1.37	0.7167
3.5	7.19	5.83	4.90	1.47	7.17	1.25	0.91094

Table No.(5):Titration of (0.025M) Amoxiciline and (0.05M) $Cu(NO_3)_2$ solution with 0.085M KOH solution. Temp.:25C^0 ,Vol.47.5ml=0.1 , μ =0.1

Ml KOH 0.085M	pH	$[KOH]_T$	$[M]_T$	$[L]_T$	$\frac{H+}{Ka}$	$[L]^-$	$\bar{n}$
0.0	3.31	0	5.26×10^{-3}	1.57×10^{-4}	54419.8	0	0
0.5	3.41	8.85×10^{-4}	5.21	1.56	43227.2	3.44×10^{-7}	0.1321
1.0	3.52	1.74×10^{-3}	5.15	1.54	33.5550	4.12	0.3058
1.5	3.68	2.60	5.10	1.53	23214.4	5.54	0.4764
2.0	3.83	3.43	5.05	1.51	16434.5	7.19	0.6482
2.5	4.02	4.25	5.00	1.50	10611.02	1.02×10^{-6}	0.8224
3.0	4.24	5.05	4.95	1.48	6393.7	1.54	1.0004
3.5	4.57	5.83	4.90	1.47	2990.5	3.00	1.1684
4.0	4.89	6.60	4.85	1.47	1431.3	5.59	1.3379
4.5	5.19	7.35	4.81	1.44	717.39	9.95	1.506
5.0	5.50	8.09	4.76	1.42	351.36	1.76×10^{-5}	1.6787

Table No,(6):Titration of (0.025M) Amoxiciline and (0.05M) $Ni(NO_3)_2$ solution with 0.085M KOH solution. Temp.:25C^0 ,Vol.47.5ml=0.1 , μ =0.1

Ml KOH 0.085M	pH	$[KOH]_T$	$[M]_T$	$[L]_T$	$\frac{H+}{Ka}$	$[L]^-$	$\bar{n}$
0.0	5.86	0	5.26×10^{-3}	1.57×10^{-4}	153.37	0	0
0.5	6.13	8.85×10^{-4}	5.21	1.56	82.36	1.8×10^{-4}	0.0974
1.0	6.40	1.74×10^{-3}	5.15	1.54	44.23	3.1	0.2419
1.5	6.61	2.60	5.10	1.53	27.27	4.7	0.3844
2.0	6.74	3.43	5.05	1.51	20.21	5.84	0.5326
2.5	6.85	4.25	5.00	1.50	15.69	6.94	0.6826
3.0	6.97	5.05	4.95	1.48	11.90	8.2	0.8514
3.5	7.08	5.83	4.90	1.47	9.24	9.72	0.9675
4.0	7.18	6.60	4.85	1.47	7.34	1.09×10^{-3}	1.1144
4.5	7.29	7.35	4.81	1.44	5.698	1.25	1.2479
5.0	7.40	8.09	4.76	1.42	4.423	1.39	1.3885

Table No.(7):Titration of (0.025M) Amoxiciline and (0.05M) $Co(NO_3)_2$ solution with 0.085M KOH solution. Temp.:25C^0 ,Vol.47.5ml=0.1 , μ =0.1

Ml KOH 0.085M	pH	$[KOH]_T$	$[M]_T$	$[L]_T$	$\frac{H+}{Ka}$	$[L]^-$	$\bar{n}$
0.0	6.05	0	5.26×10^{-3}	1.57×10^{-4}	96.77	0	0
0.5	6.46	8.85×10^{-4}	5.21	1.56	38.52	3.87×10^{-4}	0.0579
1.0	6.70	1.74×10^{-3}	5.15	1.54	22.169	6.24	0.1812
1.5	6.87	2.60	5.10	1.53	4.98	8.58	0.3082
2.0	6.99	3.43	5.05	1.51	11.369	1.04×10^{-3}	0.44259
2.5	7.08	4.25	5.00	1.50	9.241	1.17	0.5855
3.0	7.18	5.05	4.95	1.48	7.341	1.34	0.7220
3.5	7.22	5.83	4.90	1.47	6.695	1.34	0.8956
4.0	7.32	6.60	4.85	1.47	5.318	1.505	1.029
4.5	7.40	7.35	4.81	1.44	4.42	1.615	1.179
5.0	7.44	8.09	4.76	1.42	4.03	1.54	1.3463

Table No.(8):Titration of (0.025M) Amoxiciline and (0.05M) $Cr(NO_3)_3$ solution with 0.085M KOH solution. Temp.:25C^0 ,Vol.47.5ml=0.1 , μ =0.1

Ml KOH 0.085M	pH	$[KOH]_T$	$[M]_T$	$[L]_T$	$\frac{H+}{Ka}$	$[L]^-$	$\bar{n}$
0.0	4.46	0	5.26×10^{-3}	1.57×10^{-4}	3852.6	0	0
0.5	4.56	8.85×10^{-4}	5.21	1.56	3060.2	4.87×10^{-6}	0.13124
1.0	4.68	1.74×10^{-3}	5.15	1.54	2321.4	5.96	0.30135
1.5	4.85	2.60	5.10	1.53	1569.48	8.19	0.4779
2.0	5.04	3.43	5.05	1.51	1013.34	1.16×10^{-5}	0.6460
2.5	5.25	4.25	5.00	1.50	624.8	1.74	0.8178
3.0	5.48	5.05	4.95	1.48	367.9	2.68	0.9885
3.5	5.61	5.83	4.90	1.47	272.7	3.29	1.1589
4.0	5.73	6.60	4.85	1.47	206.89	3.86	1.3351
4.5	5.83	7.35	4.81	1.44	164.3	4.34	1.4995
5.0	5.86	8.09	4.76	1.42	153.37	4.03	1.6740

Table No.(9): Mesonos Metal-Amoxiline parameters calculation according to the equation LogK=αx + βy + γ ,Where K being the first association(β_1).

*X is related to the electronegatively of the ion and is obtained from the equation $(10X)^{1/2} = X^0_m + (In)^{1/2}$,Where X^0_m is the electronegativity of the metal taken from Gordy and Thomas[15] and (In) is the ionization energy (e.v)[16].

Metal ion	β_1	Metal parameter		Notes
Ag^{+}	3.25×10^{3}	*X	**Y	B_2, β_3 ,β_n ……etc Have negatives values.
Cd^{+2}	5.84×10^{3}	4.80	3.99	
Cu^{+2}	1.63×10^{9}	4.87	3.04	
Ni^{+2}	3.67×10^{4}	5.31	2.80	
Co^{+2}	3.307×10^{4}	4.73	2.82	
Cr^{+3}	3.09×10^{6}	4.39	2.59	
***Ligand parameters		8.39	2.70	
		α	0.46	
		β	1.03	
		γ	3.766	

$$**Y = 10\{\frac{In}{In+1}\}\{\frac{ri}{n^{\frac{1}{2}}}\}$$

Where(ri) is the ionic radius of metal ion and (n).is the formal charge

*** values of ligands paramers were calculated using the least squares solution of an over determined system of linear equations to derive the normal equation[17] .

تعيين ثوابت الاستقرارية لبعض المعقدات الفلزية مع (6-(2-امينو- 2-(4-هيدروكسي فينول) استيمايدو)-3,3- ثنائي مثيل-7- وكسو-4- ثايا-1- ازا- ثنائي الحلقة (0,2,3) هبتان-2- حامض الكاربوكسيلك (اموكسلين)

باسمة محسن سرحان
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استلم البحث في : 28 نيسان 2013 ، قبل البحث في :24 تموز 2013

الخلاصة

عُيِنَ ثابت التفكك للاموكسلين في درجة 25م^0 وثوابت Cr^{+3}, Co^{+2}, Ni^{+2}, Cu^{+2}, Ag^{+1} and Cd^{+2} الاستقرارية للمعقدات التي يكونها مع ان هذا الليكاند يتصرف وكانه ثنائي السن وانه يكون مع معظم المعقدات معقدات ثابتة بنسبة (1:1) مع الايونات اعلاه وحسبت معاملات الصلادة والليونة لليكاند وقد وجد ان قيمة معامل الصلادة (α) تساوي (0.46) وان معامل الليونة (β) تساوي(1.03).

الكلمات المفتاحية:-الاموكسلين، ثوابت الاستقرارية، المعقدات الفلزية