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An Absorption Spectral Analysis of the Complexation of Nd(III) with Adenosine in Presence and Absence of Zn(II) in Aqueous and Aquated Organic Solvents

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Studies on the difference in energy parameters and comparative absorption spectrophotometry involving 4*f*-4*f* transitions on Nd(III) and adenosine in presence and absence of Zn(II) have been carried out in aqueous and aquated organic solvents (50:50) like CH_3OH, dioxane, CH_3CN and DMF. Variations in the spectral energy parameters Slater-Condon (F_K) factor, Lande spin-orbit coupling constant (ξ_{4f}), nephelauxetic ratio (β), bonding parameter ($b^{1/2}$) and per cent covalency (δ) - are calculated and correlated with binding of Nd(III) with adenosine in presence and absence of Zn(II).

Key Words: Adenosine, Absorption spectra, Hypersensitive, Pseudohypersensitive, Nephelauxetic effect.

INTRODUCTION

Lanthanide coordination chemistry in solution state become a new age with the increasing use of lanthanides as probes in the exploration of the structural functions of biomolecular reactions[1-5]. This is particularly due to their ability to replace Ca(II) ions in specific manner[6]. Shah[7] studied comparative 4*f*-4*f* transition spectra of Pr(III) with lysozyme by using energy interaction parameters to explain the behaviour of binding between them. Mehta[8] also studied the mode of binding between Pr(III) and Nd(III) with lysozyme in presence of Zn(II), a soft metal ion, by employing intensity parameters. The ligand, adenosine (Fig. 1) is a naturally occurring nucleoside. It has both oxygen and nitrogen donor sites *viz.*, carbonyl, amino groups and pepetide linkage of purine nucleus and hydroxylic groups of ribose sugar of adenosine. Nucleosides and nucleotides, the building blocks of nucleic acids form a very important class of organic molecules in molecular biology.

Hard metal ions like Ca^{2+} and the soft metal ion Zn^{2+} are endogenous metal ions that have differing co-ordinating behaviour towards biological molecules. Since Nd(III) resembles Ca(II), its complexation can provide information about the coordination characteristics of diamagnetic Ca^{2+} with

biomolecules during biochemical reactions. Hence, paramagnetic lanthanides are good spectral probes for exploring the biological roles of Ca^{2+} by isomorphous substitution[9]. The present work discusses the quantitative spectral energy interaction parameters of Nd^{3+} complexes with adenosine in presence and absence of Zn^{2+} in aquated organic solvents at pH 6 and 298 K. The present work reports the sensitivity of the hypersensitive transition $^4I_{9/2} \rightarrow {}^4G_{5/2}$ and ligand mediated pseudo-hypersensitive transitions $^4I_{9/2} \rightarrow {}^4F_{3/2}$, $^4I_{9/2} \rightarrow {}^4F_{5/2}$, $^4I_{9/2} \rightarrow {}^4F_{7/2}$ and $^4I_{9/2} \rightarrow {}^4G_{7/2}$ of Nd^{3+} and uses the magnitude and variation of Slater-Condon factor (F_K, K = 2, 4, 6), Lande spin-orbit coupling (ξ_{4f}), nephelauxetic ratio (β), bonding ($b^{1/2}$) and per cent covalency (δ) parameters to discuss the bonding of Nd^{3+} with adenosine in presence and absence of Zn^{2+}.

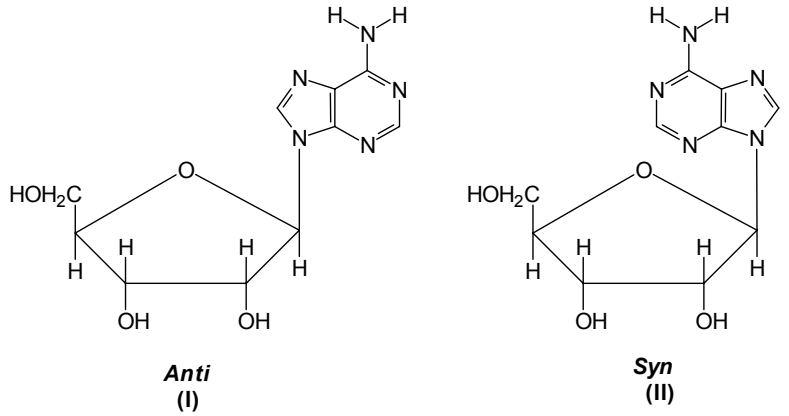

Fig. 1. *Anti* and *Syn* conformations of adenosine

EXPERIMENTAL

Neodymium(III) nitrate hexahydrate of 99.9 % purity was purchased from M/s Indian Rare Earths Ltd. and adenosine from Sisco Pvt. Ltd., Mumbai. The solvents used are CH_3CN, CH_3OH, DMF and dioxane of AR grade from E. Merk. The saturated solution of ligand and Nd(III) nitrate of 10^{-2} M were prepared in different solvents. Absorption spectra of each solution at pH 6 and at temperature 298 K were recorded on Perkin Elmer Lamda-35 UV-Vis spectrophotometer upgraded with high resolution and expansion of scale in the region 300-1100 nm by using water circulating thermostat model of HAAKE DC 10.

Nephelauxetic ratio has been regarded as a measure of covalency[10-12] and has been interpreted in terms of Slator-Condon and Racah parameters (inter electronic repulsion parameters) as well as by the ratio of the complex ion and free ion[13].

$$\beta = F_K^C / F_K^f \quad \text{or} \quad E_C^K / E_f^K \tag{1}$$

where F_K (K = 2, 4, 6) is the Slater-Condon parameter and E^k is the Racah parameter, C and f stand for complex and free ions, respectively. The bonding parameter ($b^{½}$) is inter-related to nephelauxetic effect as,

$$\beta^{½} = [(1-\beta)/2]^{½} \tag{2}$$

The electrostatic term E_o is expressed in terms of the product of Slater radial integral known as Slate Condon parameter F_k and is given by

$$E_0 = \sum_{k=0}^{k=6} K^k F_k \tag{3}$$

The Slater-Condon parameters are also known as direct integrals and are a decreasing function of K given by the relation,

$$F_I^K = \int_0^\infty \int_0^\infty \frac{r_<^K}{r_>^{K+I}} R_i^2(r_i) R_j^2(r_j) r_i^2 r_j^2 dr_i dr_j \tag{4}$$

where R is the 4*f*-radial wave function; $r_<$ and $r_>$ the radii of near and more distant electrons and i and j the ith and jth electrons under consideration. Condon and Shortley[13] redefined F^k integrals in terms of reduced integral F_k related to each other and the relation is

$$F_k = F^k / D_k \tag{5}$$

Combining relations (4) and (5), the reduced Slate Condon integral can be written as:

$$F_I^K = \frac{1}{D_K} \int_0^\infty \int_0^\infty r_<^k r_>^{k+1} R_i^2(r_i) R_j^2(r_j) T_i^2 r_j^2 dr_i dr_j \tag{6}$$

Here D_K is the denominator and F_K are coefficients of linear combination and represent the angular part of the interaction. The energy E_{s0} arising from the most important magnetic interactions, which are spin-orbit interactions, may be written as

$$E_{so} = A_{so}\xi_{4f} \tag{7}$$

where A_{so} is the angular part of spin-orbit interaction and ξ_{4f} is the radial integral and is known as Lande's parameter. By first order approximation, the energy E_j of the jth level is given by Wong[14,15] as

$$E_j(E_K, \xi_{4f}) = E_{0j}(F_K^0, \xi_{4f}) + \frac{\partial E_j}{\partial F_K} \Delta F_K + \frac{\partial E_j}{\partial \xi_{4f}} \Delta \xi_{4f} \tag{8}$$

where E_{Oj} is the zero order energy of the jth level. The value of F_k and ξ_{4f} are given by

$$F_K = F_K^0 + \Delta F_K,$$
$$\xi_{4f} = \xi_{4f}^0 + \Delta\xi_{4f}^0 \quad (9)$$

The difference between the observed E_j value and the zero order value, ΔEj, is evaluated by

$$\Delta E_j = \sum_{K=2,4,6} \frac{\partial E_j}{\partial E_K}\Delta F_K + \frac{\partial E_j}{\partial \xi_{4f}}\Delta\xi_{4f} \quad (10)$$

By using the zero order energy and partial derivatives of Nd(III) ions given by Wong[14,15], the above equation can be solved by least square technique and the value of ΔF_2, ΔF_4, ΔF_6 and $\Delta\xi_{4f}$ can be determined. The per cent covalency parameter (δ) representing the nephelauxetic effect was calculated from the relation

$$\delta = [1-\beta)/\beta] \times 100 \quad (11)$$

RESULTS AND DISCUSSION

From Fig. 2, a red shift can be observed as adenosine is added to Nd(IlI) and further longer wavelengths are observed on addition of Zn(II) in DMF. Table-1 shows the variation of the magnitude of energy interaction parameters like Slater-Condon (F_K), Lande factor (ξ_{4f}), Racah energy (E^K), nephelauxetic ratio (β), bonding parameter ($b^{1/2}$) and percentage covalency (δ) for Nd(III), Nd(III):adenosine and Nd(III):adenosine: Zn(II) in aqueous and different aquated organic solvents. Table-2 gives the computed and observed values of energies for the various transition bands and root mean square (RMS) deviation showing the correctness of the various energy parameters. There is a slight decrease in the values of F_K and ξ_{4f}, as the complexation goes on which leads to increase in the values of nephelauxetic ratio (β) and percentage covalency (δ).

For spectral studies on the structures of coordination compounds of lanthanides in solution, any evidence of the relationship between the nephelauxetic band shift and the structure is of special interest. Jorgensen and Ryan[16] noticed the dependence of the nephelauxetic effect on the coordination number and suggested that shortening in the metal-ligand distance occurs with decrease in the coordination number. To interpret the correlation, analyses of the relationships between nephelauxetic effect, geometry and energy parameters have been derived and evaluated for complex compounds. Using the angular overlap model, the value of 'n' is proportional to the nephelauxetic effect,

$$\eta = [(1-\beta^{1/2})/\beta^{1/2}] \quad (12)$$

It may be expressed as

$$\eta = \{H^{L_2}/(H_M - H_L)^2\}(S\times R)^2 N \quad (13)$$

TABLE-1
COMPUTED VALUE OF ENERGY INTERACTION SLATER-CONDON F_k (cm^{-1}), SPIN ORBIT INTERACTION ξ_{4f} (cm^{-1}), RACAH ENERGY E^k (cm^{-1}), NEPHELAUXETIC RATIO (β), BONDING PARAMETER ($b^{1/2}$) AND COVALENCY PERCENTAGE (δ) OF Nd(III), Nd(III):ADENOSINE (1:1) AND Nd(III): ADENOSINE:Zn(II) (1:1:1) SYSTEMS IN AQUEOUS AND DIFFERENT AQUATED ORGANIC SOLVENTS (50:50) AT pH 6 AND 298 K

System	F_2	F_4	F_6	ξ_{4f}	β	$b^{1/2}$	δ
Solvent-Water							
Nd(III)	328.05	48.66	5.25	957.87	1.0281	0.1186	2.7349
Nd(III):Adenosine	328.01	48.66	5.25	957.82	1.0287	0.1198	2.7905
Nd(III):Adenosine:Zn(II)	327.91	48.65	5.25	957.80	1.0295	0.1216	2.8704
Solvent-CH_3OH							
Nd(III)	330.08	48.12	5.13	928.80	1.0074	0.0607	0.7321
Nd(III):Adenosine	329.95	48.12	5.12	928.78	1.0116	0.0764	1.1526
Nd(III):Adenosine:Zn(II)	329.84	48.11	5.10	928.75	1.0128	0.0801	1.2658
Solvent-CH_3CN							
Nd(III)	330.07	48.14	5.13	928.99	1.0076	0.0616	0.7534
Nd(III):Adenosine	330.06	48.14	5.13	928.97	1.0078	0.0623	0.7707
Nd(III):Adenosine:Zn(II)	330.05	48.13	5.13	928.94	1.0079	0.0527	0.7820
Solvent-DMF							
Nd(III)	329.94	48.20	5.15	932.94	1.0106	0.0728	1.0487
Nd(III):Adenosine	329.71	48.20	5.12	932.90	1.0136	0.0825	1.3419
Nd(III):Adenosine:Zn(II)	329.60	48.12	5.06	932.83	1.0199	0.0997	1.9500
Solvent-Dioxane							
Nd(III)	329.86	48.15	5.14	931.17	1.0089	0.0665	0.8770
Nd(III):Adenosine	329.80	48.12	5.14	931.15	1.0094	0.0687	0.9346
Nd(III):Adenosine:Zn(II)	329.75	48.11	5.13	931.12	1.0110	0.0741	1.0870
Solvent-CH_3OH:CH_3CN							
Nd(III)	330.08	48.12	5.13	929.10	1.0076	0.0615	0.7515
Nd(III):Adenosine	330.07	48.11	5.13	929.08	1.0078	0.0626	0.7764
Nd(III):Adenosine:Zn(II)	330.05	48.10	5.12	929.05	1.0080	0.0633	0.7953
Solvent-CH_3OH:DMF							
Nd(III)	329.97	48.15	5.14	930.85	1.0088	0.0665	0.8729
Nd(III):Adenosine	329.75	48.14	5.13	930.80	1.0131	0.0810	1.2952
Nd(III):Adenosine:Zn(II)	329.70	48.13	5.12	930.75	1.0138	0.0831	1.3631
Solvent-CH_3OH:Dioxane							
Nd(III)	329.97	48.16	5.13	929.52	1.0079	0.0629	0.7854
Nd(III):Adenosine	329.94	48.15	5.13	929.49	1.0081	0.0637	0.8048
Nd(III):Adenosine:Zn(II)	329.93	48.14	5.12	929.47	1.0082	0.0641	0.8161
Solvent-CH_3CN:DMF							
Nd(III)	330.00	48.17	5.14	929.44	1.0081	0.0634	0.7985
Nd(III):Adenosine	329.82	48.17	5.13	929.41	1.0124	0.0788	1.2254
Nd(III):Adenosine:Zn(II)	329.76	48.15	5.10	929.40	1.0134	0.0818	1.3204
Solvent-CH_3CN:Dioxane							
Nd(III)	330.16	48.09	5.13	929.50	1.0071	0.0595	0.7037
Nd(III):Adenosine	330.12	48.07	5.13	928.48	1.0070	0.0606	0.7290
Nd(III):Adenosine:Zn(II)	330.04	48.14	5.13	928.45	1.0079	0.0628	0.7833
Solvent-DMF:Dioxane							
Nd(III)	330.01	48.17	5.13	928.39	1.0073	0.0603	0.7209
Nd(III):Adenosine	329.81	48.19	5.13	929.85	1.0126	0.0794	1.2447
Nd(III):Adenosine:Zn(II)	329.75	48.18	5.12	929.83	1.0135	0.0820	1.3268

TABLE-2
COMPUTED AND OBSERVED VALUES OF ENERGIES (cm^{-1}) AND RMS VALUES FOR Nd(III), Nd(III):ADENOSINE (1:1) AND Nd(III):ADENOSINE:Zn(II) (1:1:1) IN AQUEOUS AND DIFFEENT AQUATED ORGANIC SOLVENTS (50:50) AT pH 6 AND 298 K

System	$^4I_{9/2} \rightarrow {}^4F_{3/2}$		$^4I_{9/2} \rightarrow {}^4F_{5/2}$		$^4I_{9/2} \rightarrow {}^4F_{7/2}$		$^4I_{9/2} \rightarrow {}^4G_{5/2}$		$^4I_{9/2} \rightarrow {}^4G_{7/2}$		RMS
	E_{obs}	E_{cal}	E_{obs}	E_{cal}	E_{obs}	E_{cal}	E_{obs}	E_{cal}	E_{obs}	E_{cal}	
Solvent-Water											
Nd(III)	11477.76	11389.05	12510.32	12551.23	13422.64	13433.78	17279.81	17126.19	19105.12	19265.58	109.13
Nd(III):Adenosine	11477.10	11386.75	12509.38	12550.20	13421.92	13433.44	17275.33	17122.10	19104.02	19265.37	108.58
Nd(III):Adenosine:Zn(II)	11476.97	11382.82	12506.25	12548.56	13421.20	13432.86	17271.16	17115.30	19098.91	19265.02	111.96
Solvent-CH_3OH											
Nd(III)	11541.48	11480.89	12573.24	12602.22	13487.09	13461.37	17325.92	17286.14	19152.32	19254.95	60.86
Nd(III):Adenosine	11540.41	11466.07	12568.02	12595.93	13485.99	13459.99	17280.41	17259.09	19151.22	19253.55	59.85
Nd(III):Adenosine:Zn(II)	11536.82	11460.94	12567.39	12592.77	13478.91	13458.08	17276.53	17250.16	19150.12	19250.73	57.34
Solvent-CH_3CN											
Nd(III)	11538.42	11480.03	12572.76	12601.58	13486.18	13460.95	17328.02	17284.61	19153.05	19251.28	57.50
Nd(III):Adenosine	11538.15	11479.34	12572.45	12601.28	13485.99	13460.85	17326.82	17283.38	18152.69	19251.27	57.68
Nd(III):Adenosine:Zn(II)	11537.88	11478.86	12572.29	12601.07	13485.81	13460.78	17326.22	17282.54	19152.32	19251.23	57.27
Solvent-DMF											
Nd(III)	11547.48	11469.21	12568.66	12596.04	13479.63	13458.72	17286.08	17265.06	19153.42	19269.80	87.42
Nd(III):Adenosine	11538.15	11456.80	12560.60	12589.72	13474.36	13455.83	17275.33	17243.17	19147.92	19256.45	64.22
Nd(III):Adenosine:Zn(II)	11538.02	11429.78	12498.44	12569.57	13462.57	13441.40	17270.26	17196.70	19144.98	19254.72	60.03
Solvent-Dioxane											
Nd(III)	11539.08	11473.28	12570.08	12598.15	13478.91	13459.05	17323.52	17273.20	19143.89	19252.25	64.32
Nd(III):Adenosine	11537.62	11470.74	12566.29	12596.61	13478.18	13458.15	17322.62	17268.77	19143.15	19251.36	63.88
Nd(III):Adenosine:Zn(II)	11536.69	11465.28	12565.18	12594.30	13477.09	13457.63	17306.73	17258.82	19142.42	19250.62	62.35

System	$^4I_{9/2} \rightarrow {}^4F_{3/2}$		$^4I_{9/2} \rightarrow {}^4F_{5/2}$		$^4I_{9/2} \rightarrow {}^4F_{7/2}$		$^4I_{9/2} \rightarrow {}^4G_{5/2}$		$^4I_{9/2} \rightarrow {}^4G_{7/2}$		RMS
	E_{obs}	E_{cal}	E_{obs}	E_{cal}	E_{obs}	E_{cal}	E_{obs}	E_{cal}	E_{obs}	E_{cal}	
Solvent-CH_3OH:CH_3CN											
Nd(III)	11538.68	11480.39	12571.18	12602.12	13486.63	13461.51	17327.42	17285.15	19152.69	19251.15	58.34
Nd(III):Adenosine	11538.15	11479.44	12570.87	12601.72	13489.45	13461.40	17325.62	17283.44	19152.32	19250.89	57.76
Nd(III):Adenosine:Zn(II)	11537.88	11478.55	12601.15	12601.15	13488.91	13461.03	17325.23	17281.91	19151.95	19250.80	58.12
Solvent-CH_3OH:DMF											
Nd(III)	11537.62	11474.96	12573.40	12599.16	13481.27	13459.94	17318.12	17275.69	19150.12	19254.55	65.29
Nd(III):Adenosine	11537.49	11458.82	12564.55	12591.94	13478.54	13457.70	17277.72	17246.70	19144.62	19253.91	63.69
Nd(III):Adenosine:Zn(II)	11537.35	11455.89	12561.39	12590.38	13477.63	13456.93	17273.54	17241.55	19143.15	19251.62	58.56
Solvent-CH_3OH:Dioxane											
Nd(III)	11542.01	11477.73	12572.92	12600.11	13480.54	13459.86	17324.42	17280.87	19149.38	19152.32	57.77
Nd(III):Adenosine	11541.61	11476.73	12572.61	12599.50	13479.09	13459.46	17324.12	17279.16	19148.65	19251.35	59.81
Nd(III):Adenosine:Zn(II)	11541.35	11476.25	12572.45	12599.29	13478.91	13459.39	17323.52	17278.32	19148.29	19251.20	59.27
Solvent-CH_3CN:DMF											
Nd(III)	11539.62	11477.74	12573.08	12599.87	13480.54	13459.68	17324.12	17280.71	19152.69	19255.37	63.25
Nd(III):Adenosine	11539.35	11461.97	12568.34	12593.19	13477.45	13458.07	17279.21	17252.19	19148.29	19254.31	61.56
Nd(III):Adenosine:Zn(II)	11537.49	11458.10	12563.13	12591.26	13477.27	13457.00	17275.04	17245.33	19147.55	19252.22	57.92
Solvent-CH_3CN:Dioxane											
Nd(III)	11538.02	11482.78	12574.50	12603.68	13491.63	13462.54	17329.22	17289.23	19154.15	19251.33	57.64
Nd(III):Adenosine	11537.35	11481.51	12574.03	12602.88	13489.82	13462.02	17328.92	17287.06	19153.42	19250.43	56.37
Nd(III):Adenosine:Zn(II)	11536.29	11478.67	12572.92	12600.88	13484.72	13460.59	17328.32	17282.25	19152.32	19250.18	55.80
Solvent-DMF:Dioxane											
Nd(III)	11549.08	11480.30	12571.18	12601.08	13481.81	13459.99	17222.92	17285.45	19150.58	192256.04	63.22
Nd(III):Adenosine	11543.48	11462.17	12564.71	12592.67	13476.91	13457.33	17278.02	17252.65	19149.38	19255.30	62.67
Nd(III):Adenosine:Zn(II)	11536.42	11456.90	12562.66	12590.24	13475.82	13456.50	17273.54	17243.19	19148.29	19251.51	69.39

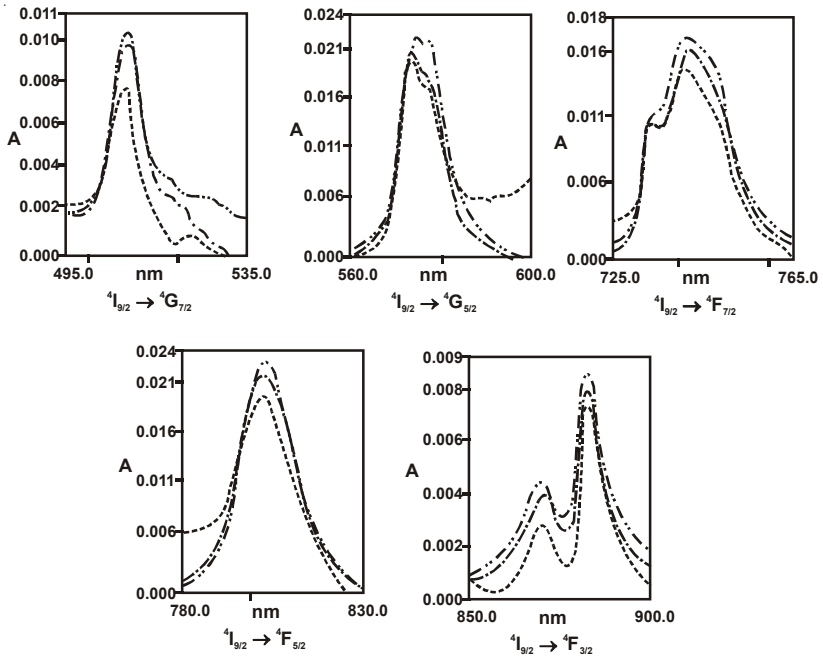

Fig. 2. Comparative absorption spectra of Nd(III) ----------, Nd(III):adenosine —·—·— and Nd(III):adenosine:Zn(II) — — — — in DMF water (50:50)

where N is the coordination number, H_M and H_L are coulomb integrals of the atomic orbital, S is the overlap integral, R is the radius of the orbit. For compounds with ligands coordinated through identical donor atoms, the first term of the RHS of (13) is a constant and (13) then becomes

$$\eta = \text{constant } (S \times R)^2 N \tag{14}$$

Eqn. 14 represents the nephelauxetic effect as a function of two variables. S×R and N which vary with changes in lanthanide-ligand distance in opposite directions. However, any variation in the value of R leads to a larger change in $(S \times R)^2$ compared to that in N. As a result, the nephelauxetic effect increases when the coordination number decreases. The (Ln-O) distance shortens in spite of the additive nature of β and decrease in the number of coordinating ligands. Variation in the value of E^K (K = 2, 4, 6); corresponds to that in the value of F^K, since they are inter-related. Misra *et al.*[17,18] observed a general decrease in the values of F_K and ξ_{4f}, parameters as compared to the corresponding parameters of the free ion.

The hypersensitive transition, $^4I_{9/2} \rightarrow G_{5/2}$ obeys the selection rule, while the ligand mediated pseudohypersensitive transitions, $^4I_{9/2} \rightarrow {}^4F_{3/2}$, $^4I_{9/2} \rightarrow {}^4F_{5/2}$, $^4I_{9/2} \rightarrow {}^4F_{7/2}$ and $^4I_{9/2} \rightarrow {}^4G_{7/2}$ of Nd(III) do not. The latter however

exhibit substantial sensitivity, reflected through the wide variation of oscillator strengths and energies with minor change in the immediate coordination environment around then even in the presence of a struturally related ligand[19,20]. Due to extremely fast water-exchange rate and very low crystal field stabilization energy, conversion from one geometry to another is very convenient and facile. Karraker[21] showed that the shape, energy and oscillator strength of hypersensitive or pseudohypersensitive transition can be correlated with coordination number and are diagnostic of the immediate coordination environment around the lanthanide ions.

Conclusion

From the present investigation it has been observed that there is possibility of the involvement of Zn^{2+} in the complexation of Nd^{3+} and adenosine, revealed by the comparative absorption spectra, which is further supported by the decreased value of the inter-electronic repulsion parameter (Slator-Condon parameter, F_K) and increased values of the nephelauxetic ratio. Further work on the evaluation of intensity parameters is going on.

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