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4f-4f Transition Spectral Analysis to Probe the Kinetics for the Complexation of Nd(III) with Guanosine and Guanosine Triphosphate (GTP)

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ABSTRACT

The interaction of Nd(III) with Guanosine and Guanosine triphosphate (GTP) in presence and absence of Ca(II) has been studied through 4*f*-4*f* transition spectra in different organic solvents. Mode of binding of Nd(III) with nucleoside (guanosine) and nucleotide (guanosine triphosphate, GTP) is interpreted considering the variations in evaluated values of intensity parameters like oscillator strength (P) and Judd Ofelt electric dipole intensity parameters (T_λ, λ=2,4,6). It is further studied through kinetics, subsequently Ea, rate constant (k) and thermodynamic parameters viz, ΔH°, ΔG°, ΔS° etc., has been evaluated from which detailed thermodynamical information of complex can be explored.

1. Introduction

Lanthanide complexes have received attention due to a wide field of application [1-4]. Considerable efforts have been made in studying metal complexes with antibacterial and antitumor activities and interactions between biological macromolecules [5, 6]. Some lanthanide complexes are biological probes for medical diagnosis and drug development [7]. Many probes for DNA analysis have been proposed [8, 9]. Recently it has been shown that some lanthanide complexes might have a potential role in the treatment of tumor cell lines [10]. They provide tremendous inspiration to many efforts in designing and synthesizing of potential anticancer and antibacterial agents because of their special electronic configuration [11-16]. Again, lanthanide co-ordination chemistry in solution state has become a new age with increasing use of lanthanides as PROBES in exploring the structural function of biomolecular reactions. It is mainly due to their isomorphous character and ability to replace Ca(II) ions in specific manner [17-21].

In the present work, we selected a nucleoside viz., guanosine and a nucleotide viz., guanosine triphosphate (GTP) for the complexation with Nd(III) in presence and absence of Ca(II) which biologically plays one of the most important role in human metabolism but being diamagnetic in nature, it is spectroscopically inert. Lanthanides, because of its paramagnetic character can be used as spectral probes for exploring the biological roles of Ca (II) through its isomorphous substitution [22-24].

4*f*-4*f* transition spectral analysis have been employed, evaluating the oscillator strength and Judd- Ofelt electronic dipole intensity parameters, T_λ (λ=2, 4, 6) for the complexation of Nd(III) with guanosine and GTP in presence and absence of Ca(II) in different aquated organic solvents. Analysis of such parameters interprets the mode of binding and nature of the complexes between guanosine/GTP with Nd (III). The formation of Nd (III): guanosine: Ca (III) and Nd (III): GTP: Ca(III) complexes is further studied through kinetic approach at different temperatures in DMF medium; where activation energy and thermodynamic parameters of complexes like ΔH°, ΔG° and ΔS° have been calculated.

2. Experimental Methods

Nd(III) nitrate hexahydrate of 99.9% purity purchased for M/s. Rear Earth Ltd. India, guanosine and GTP purchased from SISCO Pvt. Ltd. Mumbai were used. The solvents used are CH_3OH, CH_3CN, DMF and dioxane of AR grade from qualigens. The solutions of Nd(III), guanosine, guanosine tri phosphate, Ca(II) salts were prepared in different solvents with the concentration 10^{-2} M. For the present study Nd(III): ligand was kept at 1:1 molar ratio and the multimetal complexation like Nd(III):ligand:Ca(II) was also kept at 1:1:1 molar ratio. The absorption spectra were recorded on a Perkin-Elmer Lamda 35 UV-Visible spectrophotometer with high resolution and expansion of scale in a water jacketed cell holder in the region 400-1000 nm. The temperature for recording of all spectra was maintained at 298 K using water-circulating thermostat model DS-G-HAAKE.

2.1 Theoretical

The Nephelauxetic effect [25-27], a measure of covalency has been interpreted in terms of Slater Codon and Racah parameters with the measurement of free ion and complex ion [28, 29].

$$\beta = \frac{F_K^C}{F_K^f} \text{ or } \frac{E_C^K}{E_f^K} \quad (1)$$

where F_k (k=2, 4, 6) is the Slater-Condon parameter and E^k is the Racah parameters for complex and free ions respectively. The bonding parameter ($b^{1/2}$) is inter-related to Nephelauxetic effect as,

$$b^{1/2} = \left[\frac{1-\beta}{2}\right]^{1/2} \quad (2)$$

The percent covalency parameter (δ) representing the Nephelauxetic effect was given by the relation

$$\delta = \left(\frac{1-\beta}{\beta}\right) \times 100 \quad (3)$$

In the electronic transition of 4*f*-4*f*, the energy, E_{so} arises from the most important magnetic interactions, while the spin orbit interactions may be written as

$$E_{so} = A_{so}\,\xi_{4f} \quad (4)$$

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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 j^{th} level is given by Wong [30] as

$$E_j(F_K, \xi_{4f}) = E_{oj}(F_K^0, \xi_{4f}) + \frac{\partial E_j}{\partial F_K}\Delta F_K + \frac{\partial E_j}{\partial \xi_{4f}}\Delta\xi_{4f} \quad (5)$$

where E_{oj} is the zero order energy of the j^{th} level. The value of F_k and ξ_{4f} are given by

$$F_K = F_K^0 + \Delta F_K; \xi_{4f} = \xi^o{}_{4f} + \Delta\xi_{4f} \quad (6)$$

The difference between the observed Ej and the zero order values, Δ Ej is evaluated by

$$\Delta E_j = \sum_{K=2,4,6}\frac{\partial E_j}{\partial F_K}\Delta F_K + \frac{\partial E_j}{\partial \xi_{4f}}\Delta\xi_{4f} \quad (7)$$

Using the zero order energy and partial derivatives of Nd(III) ion given by Wong, the above equation can be solved by least square technique and the value of ΔF_2 and $\Delta\xi_{4f}$ can be found out. From these the value of F_2 and ξ_{4f} are evaluated using relation Eq.(6).

The calculation of the band intensities is based upon the theoretical treatment derived by Judd and Ofelt [31]. They considered the transitions are essentially electric dipole in character and the oscillator strength corresponding to the induced electric dipole transition $\psi J \rightarrow Z \psi' J'$ as given by:

$$P = \nu \sum_{\lambda=2,4,6} T_\lambda < f^n \psi J \| U^{(\lambda)} \| f^n \psi' J' >^2 \quad (8)$$

where $U^{(\lambda)}$ is the unit tensor operator of the rank which connect the initial $<f^n \psi J|$ and the final $|f^n \psi' J'>$ state through three phenomenological parameters T_λ (λ=2, 4, 6) called Judd-Ofelt parameters. These parameters are related predominantly to the radial part of the $4f^n$ wave function. The wave functions of perturbing configuration of which the nearest is $4f^{n-1}5d$.

The measured intensity of an absorption band is related to the probability (P) for the absorption of radiant energy (oscillator strength) by the expression:

$$P = (4.32 \times 10^{-9}) \int \varepsilon_{max}(\nu) d\nu \quad (9)$$

where ε_{max} is the molar extinction coefficient and ν is the energy of the band in cm^{-1}. The calculated oscillator strength can be expressed in terms of T_λ parameters as

$$\frac{P_{obs}}{\nu} = \left[U^{(2)}\right]^2 . T_2 + \left[U^{(4)}\right]^2 . T_4 + \left[U^{(6)}\right]^2 . T_6 \quad (10)$$

The transition energies and the intra configurational $U^{(\lambda)}$ matrix elements are used for the intensity analysis.

The activation energy for the complexation of Nd(III): (nucleoside/ nucleotide) with Ca(II) in DMF is calculated from the plot of log k (k= rate constant) against 1/T using Arrhenius rate equation [32, 33].

$$\log k = \log A - \frac{Ea}{2.303R}\frac{1}{T} \quad (11)$$

where A is the pre-exponential factor or frequency factor. From the slope, the activation energy (Ea) is calculated as

$$Ea = Slope \times 2.303 \times R \quad (12)$$

where R is the universal gas constant.

The thermodynamic parameters for the complexation of Nd(III): (nucleoside/ nucleotide) and Ca(II) ion is calculated using Van't Hoff plot [34] of ln k against 1/T.

$$\ln k = -\frac{\Delta H^\circ}{R}\left[\frac{1}{T}\right] + \frac{\Delta S^\circ}{R} \text{ or } \ln k = -\frac{\Delta G^\circ}{RT} \quad (13)$$

3. Results and Discussion

Lanthanides exhibit sharp spectral lines which arise from transitions among levels of f^n-configuration. The (2J + 1) fold degeneracy term of such configurations are reduced to some extent as a result of the crystal field effect. However, energy levels of rare earth ions in solid state are determined largely by spin-orbit interaction, which has a larger magnitude than that of crystal field influence. The 4*f*-electron of lanthanides is more or less protected from the influence of lattice by polarisation of $5s^2$ and $5p^6$ closed shells. Crystal field splitting effect in lanthanide ion is small, i.e., 200-300 cm^{-1} in most cases.

Hypersensitivity is observed in the absorption spectra of lanthanide trihalides in gaseous state having D_{nh} symmetry [35]. Jorgenson and Judd [36, 37] considered the phenomena of hypersensitivity and concluded that all such transitions obeyed selection rules.

Among the 4*f*-4*f* transitions of Nd(III) viz., $^4I_{9/2} \rightarrow {}^4G_{7/2}$, $^4I_{9/2} \rightarrow {}^4G_{5/2}$, $^4I_{9/2} \rightarrow {}^4F_{7/2}$, $^4I_{9/2} \rightarrow {}^4F_{5/2}$; $^4I_{9/2} \rightarrow {}^4F_{3/2}$, $^4I_{9/2} \rightarrow {}^4G_{5/2}$ and $^4G_{7/2}$ obey the selection rule and as a result, they are considered as hypersensitive transitions while others as non-hypersensitive transitions. But through experimental studies, such non-hypersensitive transitions are found to be quite sensitive towards even minor changes in the co-ordination environment around lanthanide induced by design, density, chelating capability and co-ordinating power of donor sites of ligand and referred these transitions as pseudo-hypersensitive transitions, correlating with the coordination number of lanthanide ions. It is quite challenging to note that the present study is carried out only in solution state because when we focus on human metabolism and role of Ca(II) as well as guanosine and guanosine triphosphate (GTP), the most valid information can be gathered from studies particularly involving the solution state.

Table 1 Observed and computed values of oscillator strength (Px10^6) and Judd Ofelt (T_λx10) parameters for Nd(III), Nd(III):L, Nd(III):L:Ca(II),(1:1:1) (L: guanosine, GTP) in different aquated organic solvents

System	$^4F_{3/2}$ Pobs (Pcal)	$^4F_{5/2}$ Pobs (Pcal)	$^4F_{7/2}$ Pobs (Pcal)	$^4G_{5/2}$ Pobs (Pcal)	$^4G_{7/2}$ Pobs (Pcal)	T_2	T_4	T_6
1. Solvent: CH_3CN								
L: Guanosine								
Nd(III)	0.2886	3.6876	2.5468	2.0609	0.4940	20.6427	0.5478	42.1824
	(0.6921)	(3.6653)	(2.4528)	(2.1512)	(0.2841)			
Nd(III):L	0.9119	4.6065	3.0248	2.5535	0.5046	32.2733	-9.5172	46.3833
	(0.7406)	(4.6271)	(2.6097)	(2.2394)	(0.3403)			
Nd(III):L:Ca(II)	0.9256	4.6834	3.6743	2.6187	0.5327	32.7205	-1.6376	46.4734
	(0.7945)	(4.6795)	(2.6376)	(2.3171)	(0.3968)			
L: GTP								
Nd(III)	0.5381	1.8735	2.4022	1.4657	0.3743	10.2071	0.5087	28.6894
	(0.5395)	(1.8734)	(2.2822)	(1.0017)	(0.2645)			
Nd(III):L	0.1986	1.8820	2.8214	1.5373	0.4407	11.3366	-3.7033	29.8169
	(0.6000)	(1.8763)	(2.6905)	(1.3816)	(0.2901)			
Nd(III):L:Ca(II)	0.2499	1.9414	2.8945	1.9481	0.4816	11.5374	-4.031	29.9194
	(0.6121)	(1.9379)	(2.7664)	(1.4906)	(0.3458)			
2. Solvent: DMF								
L: Guanosine								
Nd(III)	0.5850	3.5905	2.7646	2.0994	0.4217	21.7099	-1.0212	45.7634
	(0.6780)	(3.5854)	(2.6208)	(2.2588)	(0.262)			
Nd(III):L	1.0386	4.7499	2.7874	2.1661	0.4681	27.9639	2.1416	46.4034
	(0.8348)	(4.7612)	(2.6377)	(2.3326)	(0.3935)			
Nd(III):L:Ca(II)	1.0529	4.7598	2.7986	2.1818	0.4728	27.9928	-1.7828	46.5445
	(0.8498)	(4.7705)	(2.6392)	(2.3483)	(0.3944)			

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L: GTP								
Nd(III)	0.9958	3.018	3.0001	2.3229	0.4168	13.5844	0.9749	30.4859
	(0.797)	(3.0286)	(2.847)	(2.5045)	(0.3385)			
Nd(III):L	1.5057	3.7321	3.3151	2.5188	0.4608	21.4117	2.569	40.6115
	(0.8118)	(3.7373)	(2.8534)	(2.6155)	(0.3546)			
Nd(III):L:Ca(II)	1.5563	3.8030	3.8718	2.5645	0.4713	21.9706	-0.4922	41.4387
	(0.8247)	(3.8048)	(2.9559)	(2.6975)	(0.3745)			
3. Solvent: CH_3OH								
L: Guanosine								
Nd(III)	0.6289	3.6364	2.5345	1.9547	0.4106	21.5429	0.4067	41.8123
	(0.6775)	(3.6338)	(2.4020)	(2.1029)	(0.2748)			
Nd(III):L	0.7317	3.6708	2.5367	1.9724	0.4273	22.5339	0.9776	41.9414
	(0.6949)	(3.6729)	(2.4066)	(2.1214)	(0.2901)			
Nd(III):L:Ca(II)	0.7400	3.6871	2.5471	1.9751	0.4296	22.9819	1.1620	42.9200
	(0.6950)	(3.6846)	(2.4146)	(2.1309)	(0.2992)			
L: GTP								
Nd(III)	0.6938	2.1485	2.6684	2.0667	0.3744	10.9506	0.239	26.1692
	(0.5969)	(2.1539)	(2.5384)	(2.2180)	(0.2856)			
Nd(III):L	0.7043	2.3431	2.7030	2.3890	0.4719	11.8368	-2.2766	29.0255
	(0.6066)	(2.3418)	(2.5626)	(2.4339)	(0.3029)			
Nd(III):L:Ca(II)	0.7215	2.3467	0.7382	2.4026	0.5731	12.8357	-2.2614	29.4856
	(0. 7159)	(2.3456)	(2.5891)	(2.4575)	(0.3162)			
4. Solvent: Dioxane								
L: Guanosine								
Nd(III)	0.7806	3.6971	2.4676	2.0490	0.4264	20.6013	3.4583	41.0407
	(0.7513)	(3.6988)	(2.3750)	(2.1551)	(0.3510)			
Nd(III):L	0.7891	3.7681	2.5439	2.1812	0.4969	22.2269	0.8112	42.0305
	(0.7894)	(3.7692)	(2.4166)	(2.1763)	(0.3670)			
Nd(III):L:Ca(II)	0.7934	3.8517	2.5520	2.1998	0.4982	22.2934	1.9098	42.4644
	(0.7972)	(3.8543)	(2.4206)	(2.1807)	(0.3826)			
L: GTP								
Nd(III)	0.7405	2.1452	2.7177	2.1366	0.4332	10.1033	2.0037	31.7166
	(0.6467)	(2.1504)	(2.5793)	(2.2974)	(0.3359)			
Nd(III):L	0.7921	2.3028	2.8976	2.4580	0.4513	12.7974	-2.547	32.1699
	(0.6748)	(2.2927)	(2.8324)	(2.5240)	(0.3557)			
Nd(III):L:Ca(II)	0.7961	2.3971	2.9038	2.7623	0.4963	13.2457	-3.1994	38.7087
	(0.0685)	(2.3175)	(2.8472)	(2.6091)	(0.3891)			

Table 2 Observed and calculated oscillator strengths (Px10^6) and Judd Ofelt parameter (T_λ, λ=2, 4, 6 x 10^{10} cm^{-1}) parameters for Nd(III):guanosine:Ca(II) complex at (a) 298 K (25 °C) at different time (h), (b) at 303 K (30 °C) at different time (h), (c) at 308 K (35 °C) at different time (h), (d) at 313 K (40 °C) at different time (h), (e) at 318 K (45 °C) at different time (h)

a) 298 K (25 °C)

Time	$^4I_{9/2}\rightarrow{}^4F_{3/2}$		$^4I_{9/2}\rightarrow{}^4F_{5/2}$		$^4I_{9/2}\rightarrow{}^4F_{7/2}$		$^4I_{9/2}\rightarrow{}^4G_{5/2}$		$^4I_{9/2}\rightarrow{}^4G_{7/2}$		T_2	T_4	T_6
(h)	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}			
0	0.4712	0.382	1.3798	1.6974	1.4001	1.3239	1.1422	1.0032	0.2791	0.2090	0.6570	0.3217	2.2839
2	0.4794	0.3876	1.4151	1.7145	1.4105	1.3358	1.1561	1.0289	0.2807	0.2130	0.6755	0.3295	2.3041
4	0.4836	0.3828	1.4359	1.7440	1.4471	1.3674	1.1718	1.0441	0.2815	0.2119	0.6980	0.3049	2.3618
6	0.4862	0.3813	1.4673	1.7844	1.4882	1.4069	1.1994	1.0668	0.2824	0.2124	0.7249	0.2843	2.4331
8	0.5356	0.4038	1.5091	1.8186	1.5032	1.4226	1.2237	1.0985	0.2853	0.2235	0.7314	0.3298	2.4560
10	0.5703	0.4133	1.6061	1.8905	1.5851	1.4838	1.2518	1.1697	0.3044	0.2319	0.7925	0.3261	2.5637
12	0.5801	0.4275	1.6236	1.9309	1.6049	1.5112	1.2898	1.1823	0.3064	0.2382	0.7919	0.3480	2.6095
14	0.5803	0.4331	1.6370	1.9570	1.6235	1.5319	1.3112	1.1918	0.3094	0.2407	0.7974	0.3514	2.6455
16	0.5982	0.4487	1.6537	1.9814	1.6358	1.5435	1.3279	1.2041	0.3172	0.2476	0.7936	0.3824	2.6629
18	0.5997	0.4458	1.6751	1.9911	1.6517	1.5548	1.3297	1.2198	0.3194	0.2475	0.8119	0.3711	2.6837
20	0.6013	0.4513	1.6959	2.0112	1.6668	1.5697	1.3443	1.2347	0.3241	0.2507	0.8212	0.3777	2.7092
22	0.6232	0.4615	1.7078	2.0338	1.6841	1.5836	1.3562	1.2439	0.3291	0.2553	0.8203	0.3958	2.7316
24	0.6460	0.4575	1.7279	2.0576	1.7216	1.6094	1.3594	1.2595	0.3307	0.2544	0.8420	0.3747	2.7792
26	0.6477	0.4803	1.7624	2.0961	1.7306	1.6288	1.3996	1.2838	0.3394	0.2651	0.8419	0.4199	2.8085
28	0.6551	0.4809	1.7821	2.1062	1.7414	1.6376	1.4045	1.2983	0.3401	0.2663	0.8552	0.4177	2.8243
30	0.6595	0.4822	1.8236	2.1264	1.7618	1.6561	1.4171	1.3285	0.3427	0.2688	0.8831	0.4127	2.8570
32	0.6696	0.4819	1.8603	2.1733	1.8084	1.7007	1.4487	1.3552	0.3429	0.2701	0.9129	0.3928	2.9373
34	0.6747	0.4861	1.8715	2.1871	1.8170	1.7109	1.4605	1.3634	0.3437	0.2722	0.9172	0.3979	2.9545

b) 303 K (30 °C)

Time	$^4I_{9/2}\rightarrow{}^4F_{3/2}$		$^4I_{9/2}\rightarrow{}^4F_{5/2}$		$^4I_{9/2}\rightarrow{}^4F_{7/2}$		$^4I_{9/2}\rightarrow{}^4G_{5/2}$		$^4I_{9/2}\rightarrow{}^4G_{7/2}$		T_2	T_4	T_6
(h)	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}			
0	0.4713	0.3897	1.3805	1.6899	1.3894	1.3110	1.1342	1.0035	0.2868	0.2122	0.6478	0.3448	2.2593
2	0.4980	0.3996	1.4134	1.7169	1.4159	1.3292	1.1438	1.0282	0.2952	0.2178	0.6616	0.3599	2.2899
4	0.5661	0.4900	1.5117	1.7810	1.4201	1.3227	1.1778	1.0998	0.3555	0.2590	0.6302	0.5718	2.2577
6	0.5676	0.5156	1.5179	1.8099	1.4229	1.3311	1.2056	1.1038	0.3703	0.2693	0.6079	0.6277	2.2668
8	0.5707	0.5274	1.5399	1.8308	1.4294	1.3420	1.2261	1.1195	0.3750	0.2750	0.6108	0.6506	2.2837
10	0.5768	0.5125	1.6503	1.8748	1.5021	1.3950	1.2356	1.1998	0.3831	0.2746	0.7047	0.5935	2.3823
12	0.5833	0.5199	1.6716	1.8970	1.5142	1.4104	1.2553	1.2152	0.3848	0.2786	0.7124	0.6046	2.4080
14	0.5916	0.5416	1.6740	1.9373	1.5267	1.4325	1.2958	1.2168	0.3907	0.2868	0.6940	0.6449	2.4430
16	0.6260	0.5457	1.7064	1.9701	1.5664	1.4606	1.3053	1.2413	0.3952	0.2899	0.7151	0.6424	2.4924
18	0.6292	0.5406	1.7367	1.9666	1.5767	1.4610	1.2916	1.2636	0.3999	0.2897	0.7402	0.6307	2.4943

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20	0.6305	0.5576	1.7442	1.9955	1.5821	1.4758	1.3235	1.2686	0.4046	0.2965	0.7287	0.6638	2.5170
22	0.6385	0.5817	1.7595	2.0328	1.5919	1.4932	1.3593	1.2793	0.4146	0.3066	0.7152	0.7122	2.5426
24	0.6465	0.6019	1.7762	2.0829	1.6166	1.5255	1.4047	1.2913	0.4219	0.3151	0.7084	0.7461	2.5961
26	0.6497	0.5969	1.8342	2.0862	1.6451	1.5326	1.3829	1.3334	0.4379	0.3162	0.7523	0.7309	2.6098
28	0.6509	0.6268	1.8674	2.1221	1.6625	1.5444	1.4032	1.3567	0.4667	0.3296	0.7433	0.7950	2.6242
30	0.6510	0.6039	1.8707	2.1828	1.7597	1.6188	1.4213	1.3595	0.4718	0.3199	0.7786	0.7108	2.7627
32	0.6537	0.6025	1.8832	2.2202	1.8041	1.6553	1.4396	1.3687	0.4780	0.3194	0.7929	0.6918	2.8288
34	0.6682	0.6205	1.8941	2.2421	1.8084	1.6626	1.4587	1.3767	0.4854	0.3271	0.7819	0.7308	2.8377

(c) 308 K (35 °C)

Time	$^4I_{9/2}\rightarrow{}^4F_{3/2}$		$^4I_{9/2}\rightarrow{}^4F_{5/2}$		$^4I_{9/2}\rightarrow{}^4F_{7/2}$		$^4I_{9/2}\rightarrow{}^4G_{5/2}$		$^4I_{9/2}\rightarrow{}^4G_{7/2}$		T_2	T_4	T_6
(h)	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}			
0	0.4291	0.3469	1.3812	1.6575	1.3906	1.3119	1.1112	1.0034	0.2672	0.1955	0.6924	0.2459	2.2711
2	0.4761	0.3657	1.4214	1.6979	1.4161	1.3366	1.1385	1.0338	0.2693	0.2048	0.7032	0.2782	2.3110
4	0.4871	0.3664	1.4472	1.7273	1.4495	1.3637	1.1524	1.0528	0.2733	0.2061	0.7226	0.2686	2.3595
6	0.4941	0.3771	1.4852	1.7579	1.4690	1.3851	1.1770	1.0803	0.2790	0.2121	0.7387	0.2840	2.3952
8	0.5085	0.3739	1.5162	1.7573	1.4794	1.3868	1.1662	1.1033	0.2808	0.2127	0.7627	0.2760	2.3990
10	0.5377	0.3608	1.6020	1.8151	1.5676	1.4514	1.1802	1.1667	0.2866	0.2114	0.8411	0.2181	2.5182
12	0.5510	0.3710	1.6231	1.8726	1.6232	1.4982	1.2119	1.1822	0.3000	0.2160	0.8498	0.2221	2.5997
14	0.5639	0.3714	1.6416	1.8729	1.6276	1.4982	1.2067	1.1960	0.3007	0.2173	0.8617	0.2231	2.5995
16	0.5684	0.3970	1.6578	1.9211	1.6345	1.5248	1.2645	1.2073	0.3021	0.2278	0.8482	0.2711	2.6415
18	0.5774	0.4041	1.6777	1.9388	1.6458	1.5363	1.2774	1.2218	0.3053	0.2315	0.8553	0.2823	2.6607
20	0.5777	0.4251	1.6876	1.9597	1.6506	1.5407	1.2928	1.2284	0.3218	0.2402	0.8399	0.3288	2.6637
22	0.5791	0.4355	1.7199	1.9791	1.6665	1.5512	1.3011	1.2517	0.3356	0.2461	0.8507	0.3492	2.6802
24	0.5799	0.4445	1.7243	1.9981	1.6814	1.5624	1.3109	1.2547	0.3459	0.2497	0.8455	0.3650	2.6985
26	0.5887	0.4579	1.7712	2.0315	1.7036	1.5842	1.3348	1.2886	0.3555	0.2573	0.8642	0.3871	2.7342
28	0.6031	0.4840	1.7988	2.0891	1.7242	1.6194	1.3932	1.3085	0.3593	0.2686	0.8591	0.4323	2.7915
30	0.6078	0.4874	1.8149	2.1300	1.7591	1.6556	1.4244	1.3200	0.3603	0.2703	0.8703	0.4248	2.8557
32	0.6196	0.4899	1.8593	2.1526	1.7845	1.6752	1.4340	1.3525	0.3654	0.2735	0.8990	0.4224	2.8903
34	0.6257	0.4891	1.8892	2.1791	1.8131	1.7010	1.4501	1.3744	0.3667	0.2745	0.9225	0.4097	2.9366

(d) 313 K (40 °C)

Time	$^4I_{9/2}\rightarrow{}^4F_{3/2}$		$^4I_{9/2}\rightarrow{}^4F_{5/2}$		$^4I_{9/2}\rightarrow{}^4F_{7/2}$		$^4I_{9/2}\rightarrow{}^4G_{5/2}$		$^4I_{9/2}\rightarrow{}^4G_{7/2}$		T_2	T_4	T_6
(h)	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}			
0	0.4381	0.3553	1.3831	1.6623	1.4006	1.3106	1.1018	1.0050	0.2811	0.1988	0.6852	0.2654	2.2670
2	0.4819	0.3676	1.4200	1.7028	1.4345	1.3399	1.1248	1.0332	0.2828	0.2054	0.7011	0.2812	2.3167
4	0.4889	0.3738	1.4528	1.7176	1.4408	1.3493	1.1391	1.0569	0.2835	0.2095	0.7170	0.2914	2.3323
6	0.5131	0.3644	1.4729	1.7230	1.4699	1.3613	1.1225	1.0726	0.2853	0.2069	0.7421	0.2650	2.3557
8	0.5207	0.3784	1.5218	1.7543	1.4811	1.3806	1.1545	1.1079	0.2879	0.2149	0.7612	0.2894	2.3870
10	0.5223	0.3813	1.6042	1.8476	1.5633	1.4671	1.2278	1.1672	0.2892	0.2193	0.8220	0.2590	2.5418
12	0.5327	0.3846	1.6236	1.8740	1.5865	1.4896	1.2458	1.1815	0.2901	0.2214	0.8341	0.2568	2.5814
14	0.5550	0.3955	1.6336	1.9032	1.6039	1.5091	1.2689	1.1894	0.2905	0.2260	0.8321	0.2740	2.6140
16	0.5680	0.4089	1.6580	1.9366	1.6209	1.5306	1.2979	1.2071	0.2939	0.2325	0.8363	0.2964	2.6494
18	0.5691	0.4190	1.6860	1.9579	1.6250	1.5433	1.3236	1.2272	0.2943	0.2378	0.8452	0.3143	2.6700
20	0.5720	0.4245	1.6980	1.9734	1.6334	1.5540	1.3376	1.2359	0.2959	0.2404	0.8487	0.3221	2.6881
22	0.5763	0.4246	1.7100	2.0071	1.6693	1.5861	1.3578	1.2446	0.2991	0.2407	0.8604	0.3086	2.7455
24	0.6006	0.4352	1.7389	2.0229	1.6779	1.5934	1.3670	1.2663	0.3018	0.2463	0.8699	0.3293	2.7564
26	0.6109	0.4347	1.7681	2.0578	1.7141	1.6265	1.3886	1.2877	0.3026	0.2474	0.8930	0.3153	2.8155
28	0.6203	0.4450	1.7952	2.0825	1.7271	1.6422	1.4096	1.3074	0.3061	0.2528	0.9018	0.3324	2.8415
30	0.6214	0.4491	1.8136	2.1241	1.7691	1.6788	1.4338	1.3206	0.3151	0.2548	0.9139	0.3261	2.9061
32	0.6396	0.4617	1.8670	2.1552	1.7850	1.6988	1.4609	1.3595	0.3171	0.2626	0.9378	0.3470	2.9392
34	0.6494	0.4693	1.8988	2.1690	1.7955	1.7062	1.4673	1.3828	0.3236	0.2672	0.9516	0.3612	2.9509

(e) 318 K (45 °C)

Time	$^4I_{9/2}\rightarrow{}^4F_{3/2}$		$^4I_{9/2}\rightarrow{}^4F_{5/2}$		$^4I_{9/2}\rightarrow{}^4F_{7/2}$		$^4I_{9/2}\rightarrow{}^4G_{5/2}$		$^4I_{9/2}\rightarrow{}^4G_{7/2}$		T_2	T_4	T_6
(h)	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}	P_{obs}	P_{cal}			
0	0.4888	0.3423	1.4005	1.6497	1.3967	1.3084	1.0925	1.0199	0.2543	0.1948	0.7120	0.2359	2.2663
2	0.4924	0.3584	1.4313	1.6863	1.4143	1.3313	1.1253	1.0420	0.2620	0.2026	0.7177	0.2634	2.3034
4	0.4990	0.3605	1.4631	1.7142	1.4410	1.3560	1.1433	1.0652	0.2646	0.2049	0.7391	0.2579	2.3474
6	0.5166	0.3803	1.4827	1.7591	1.4583	1.3838	1.1876	1.0795	0.2660	0.2134	0.7345	0.2920	2.3927
8	0.5236	0.3810	1.5108	1.7855	1.4837	1.4082	1.2055	1.1000	0.2666	0.2149	0.7549	0.2837	2.4363
10	0.5294	0.3647	1.6047	1.8419	1.5766	1.4739	1.2156	1.1683	0.2804	0.2128	0.8412	0.2179	2.5581
12	0.5386	0.3732	1.6231	1.8594	1.5836	1.4844	1.2323	1.1818	0.2815	0.2170	0.8458	0.2327	2.5749
14	0.5590	0.3845	1.6500	1.8796	1.5957	1.4950	1.2448	1.2017	0.2862	0.2228	0.8530	0.2544	2.5915
16	0.5641	0.4014	1.6693	1.9174	1.6060	1.5182	1.2870	1.2154	0.2868	0.2303	0.8503	0.2839	2.6292
18	0.5757	0.3891	1.6848	1.9223	1.6357	1.5320	1.2719	1.2273	0.2890	0.2261	0.8755	0.2495	2.6568
20	0.5840	0.4193	1.7052	1.9636	1.6303	1.5486	1.3272	1.2417	0.2906	0.2390	0.8587	0.3126	2.6798
22	0.5907	0.4338	1.7135	1.9998	1.6496	1.5723	1.3589	1.2476	0.2962	0.2447	0.8519	0.3356	2.7191
24	0.5966	0.4098	1.7343	2.0047	1.6953	1.5949	1.3355	1.2634	0.2980	0.2362	0.8939	0.2705	2.7651
26	0.6076	0.4295	1.7700	2.0554	1.7153	1.6282	1.3875	1.2891	0.2993	0.2455	0.9001	0.3024	2.8202
28	0.6094	0.4622	1.7964	2.0986	1.7251	1.6446	1.4269	1.3075	0.3189	0.2596	0.8842	0.3713	2.8422
30	0.6133	0.4799	1.8378	2.1303	1.7279	1.6616	1.4667	1.3372	0.3195	0.2686	0.8943	0.4049	2.8688
32	0.6150	0.4822	1.8717	2.1791	1.7747	1.7062	1.4997	1.3616	0.3245	0.2707	0.9192	0.3913	2.9482
34	0.6272	0.5009	1.8936	2.2120	1.7941	1.7236	1.5214	1.3774	0.3379	0.2791	0.9159	0.4272	2.9751

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Absolute values of oscillator strength and Judd- Ofelt intensity parameters T_λ (λ=2, 4, 6) is determined under different experimental conditions involving interactions of Nd(III) and guanosine/GTP. Noticeable increase in magnitudes of Judd Ofelt parameters related to the binding of ligands (guanosine/GTP) in solution, shown in Table 1. Intensification becomes more when Ca(II) ion is added to binary mixture of Nd(III) and ligand (guanosine/ GTP), and signifies binding of Ca(II) to complex formed. Value of T_2 is found to have significant changes, showing the changes in symmetry properties of complex species. Oscillator strength (P) values of Nd(III):guanosine/GTP complexes are shown in Table 1 showing significant variations. These changes provide evidence of ligand involvement (guanosine/GTP) in the inner sphere co-ordination of Nd(III).

Observations from the tables are supported by comparative absorption spectra shown in Figs. 1 and 2, where there is significant change when Nd(III) interacts with guanosine/GTP in solution. Comparative absorption spectra of Nd(III), Nd(III): guanosine and Nd(III): guanosine: Ca(II) in DMF vividly shows that addition of guanosine to Nd(III) results in enhancement in oscillator strength values of different 4*f*-4*f* transitions (Fig. 1). Consequently, noticeable increase is observed in magnitudes of Judd Ofelt T_λ (λ =2,4,6) parameters where T_4 is neglected because of its –ve value. Such increase in values of oscillator strengths and T_λ- parameters is more when Ca(II) is added to Nd(III): guanosine. This is due to the involvement of Ca(II) to other ligating site of guanosine. Same observation is made when guanosine is replaced by GTP as shown in Fig. 2.

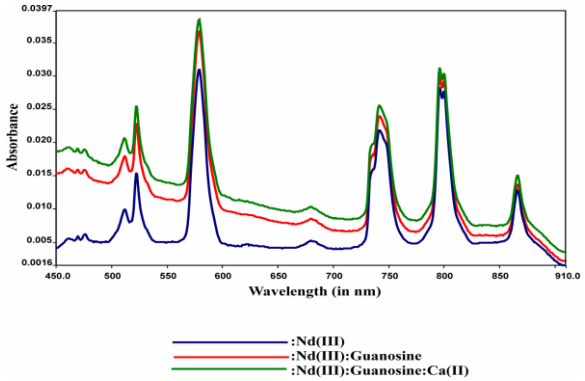

Fig. 1 Comparative absorption spectra of Nd(III), Nd(III):guanosine, Nd(III): guanosine:Ca(II) in DMF solvent

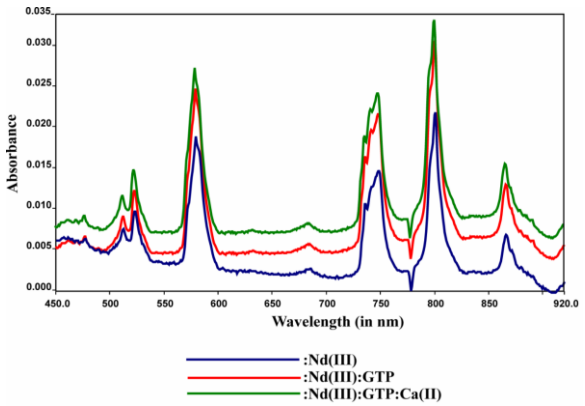

Fig. 2 Comparative absorption spectra of Nd(III), Nd(III):GTP, Nd(III):GTP:Ca(II) in DMF solvent

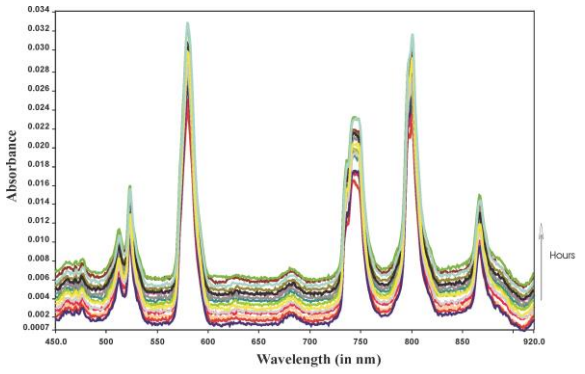

Fig. 3 Absorption spectra of Nd(III):guanosine:Ca(II) in DMF at different hours at 298 K (25 °C)

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In our series of studies, intensity of 4*f*-4*f* transitions showed substantial increase with time (Table 2), hence absorption spectral analysis of these transitions can be used to explore the kinetics of complexes formed. Comparative absorption spectra of Nd(III): guanosine: Ca(II) at different time intervals on five different temperatures viz., 298 K, 303 K, 308 K, 313 K and 318 K respectively, was analysed as shown in Fig. 3 at 298 K. Plots of oscillator strength of $^4I_{9/2} \rightarrow {}^4G_{5/2}$ transition of Nd(III) complex formation versus time (in hr) at each temperature was recorded as shown in Fig. 4.

Observed values of rate constant (k) were evaluated in terms of complex formed during the progress of reaction from tables shown above. Consequently, activation energy (Ea), rate constant (k), ΔH°, ΔS° and ΔG° of complexation of Nd (III): guanosine with Ca (II) in DMF has been evaluated. Values of rate constants at different temperatures i.e., 298 K, 303 K, 308 K, 313 K and 318 K are given in Table 3.

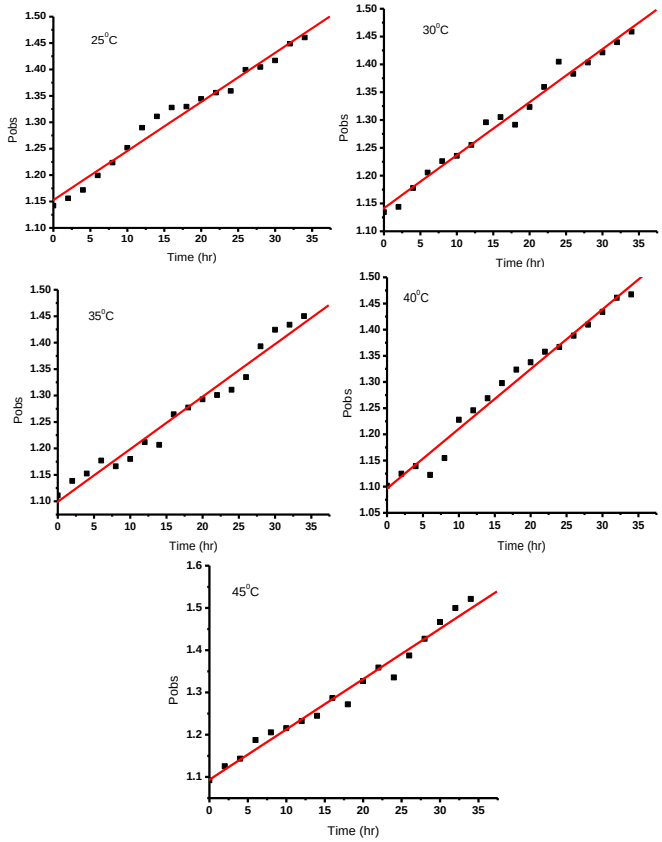

Fig. 4 Plot of P_{obs} and time (hr) for the $^4I_{9/2} \rightarrow {}^4G_{5/2}$ transition of Nd(III): guanosine and Ca(II) at 298 K, 303 K, 308 K, 313 K and 318 K respectively

Table 3 Rate Constants and thermodynamic parameters for the complexation of Nd(III): guanosine: Ca(II) at different temperatures

Temp(K)	Rate (k) Mol $L^{-1}S^{-1}$x 10^{-6}	ΔH° (kJmol^{-1})	ΔS° (JK^{-1}mol^{-1})	ΔG° (kJmol^{-1})
298	2.5833		0.0079	-2.352
303	2.6472		0.0081	-2.453
308	2.7611	0.010601	0.0085	-2.601
313	3.1694		0.0096	-3.002
318	3.3083		0.0099	-3.164

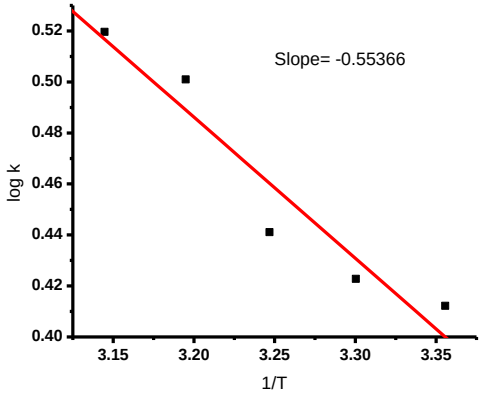

Fig. 5 Plot of lnk versus (1/T) x 10^{-3} for the complexation of Nd(III): guanosine: Ca(II) in DMF medium

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Activation energy (Ea) and thermodynamic parameters are evaluated from Van't Hoff 's plot of ln K against 1/T (Fig. 5). Kinetic studies of Nd (III): GTP with Ca(II) ions in DMF medium have been studied through same approach following the evaluation of thermodynamic parameters as shown in Table 4.

Table 4 Rate Constants and thermodynamic parameters for the complexation of Nd(III): GTP: Ca(II) at different temperatures

Temp(K)	Rate (k) Mol $L^{-1}S^{-1}x\ 10^{-6}$	ΔH° ($kJmol^{-1}$)	ΔS° ($JK^{-1}mol^{-1}$)	ΔG° ($kJmol^{-1}$)
298	2.0861		0.0062	-1.822
303	2.1278		0.0063	-1.903
308	2.2611	0.006775	0.0068	-2.090
313	2.3306		0.0070	-2.202
318	2.4722		0.0075	-2.393

4. Conclusion

From the present systematic investigation, it has been found that 4*f*-4*f* transition spectra can study the nature of binding of Nd (III) with biologically important ligands, Guanosine and GTP alongwith Ca(II) during their complexation. Increased values of oscillator strength (P) and Judd Ofelt parameters T_λ (λ= 2, 4, 6) shows strong binding of metal ion Nd(III) with Guanosine and GTP. From kinetic analysis of their complexation, it has been clearly seen that rate increases with increase in temperature, for which values of activation energy are evaluated. -ve values of ΔG° signifies that reaction is favoured in solution. The +ve values of enthalpy change ΔH° and entropy change ΔS° indicates that complexation reaction is endothermic as well as entropy increasing process. Since $T\Delta S^o > \Delta H^o$, the co-ordination reaction is entropy-driven spontaneous process.

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