Comparative Absorption Spectroscopy Involving 4f-4f Transitions to Explore the Interaction between the Pr(III) and Uracil in Presence and Absence of Zn(II)

Ch. Victory Devi and N. Rajmuhon Singh

Abstract—The interaction of Pr(III) with uracil has been explored in presence and absence of Zn(II) using comparative absorption spectroscopy involving 4f-4f transitions in different aquated organic solvents. The change in coordination in different medium is observed from the absorption intensity analysis when Pr(III) interacts with uracil. The interaction induced substantial changes in the intensities of 4f-4f bands and their perturbation was reflected through oscillator strength and Judd-Ofelt intensity (T_λ) parameters. Intensification of these bands became more prominent in presence of Zn(II) suggesting the stimulative effect of Zn(II) towards the complexation of Pr(III) with uracil. It is also suggested that the changes in the oscillator strengths of different 4f-4f bands and Judd-Ofelt intensity parameters can be used to predict in vivo intracellular complexation of uracil with Ca(II) through Pr(III)-uracil absorption spectral analysis studies in vitro as both Pr(III) and Ca(II) have unique similarity in their coordination behavior. Other parameters namely energy interaction parameters like Slator-Condon (Fk's), spin-orbit Lande (E4f), Racah parameter (Ek), nephelauxetic effect (β), bonding (b1/2) and percent covalency (δ) parameters are computed to correlate their simultaneous binding of metal ions with uracil.

Index Terms—Comparative Absorption, 4f-4f Transition, Nephelauxetic Effect, Oscillator Strength, Pr(III).

I. INTRODUCTION

The unique similarity between the lanthanides and calcium in terms of size and the coordination preference for higher coordination numbers, the former has been used as probes in biochemical reactions and for structural studies of biomolecule compounds involving Ca(II) [1-4]. The paramagnetic nature and the presence of 4f electrons have given lanthanide ions properties which can be utilized as an absorption spectral probe. Lanthanides are also finding selective applications in sol gel [5] process, which is found to be one of the most effective and extensively used methods for generation of multicompound novel materials like metal oxide and thin films. Recently, there has been renewed interest in the absorption of 4f-4f transitions and their intensity analysis to probe the finer details of structure in lanthanide complexes. Misra et al [6], made a detail studies on the interaction of Pr(III) and Nd(III) with β -diketones and

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diols using the comparative absorption spectrophotometry and reported a relation between the magnitude and variation of intensity parameters and structural and compositional changes in these complexes. David et al [7,8] have also used the electric dipole intensity and variation in the spectral parameters to explore the interaction between the Pr(III) and Nd(III) with glutathione. Only a few references are available on lanthanide-organic metabolite interactions [9] and studies on lanthanide-nucleic acid interaction are still fewer [10,11]. No attempt has been made to explore the relative and preferential binding capability of different binding sites of uracil in presence of hard metal Pr(III) and soft metal Zn(II). We thought it worthwhile to utilize absorption difference and comparative absorption spectrophotometry along with variation and magnitude of various spectral parameters to explore the simultaneous coordination of Pr(III) and Zn(II) to uracil. Zn(II), an endogeneous metal occur intracellularly and is known to form very stable complex in vivo. So, Zn(II) may involve heterobimetallic simultaneous complexation with uracil.

In the present study, the interaction of uracil with Pr(III) in presence and absence of Zn(II) is followed by selecting different aquated organic solvents and the corresponding changes in oscillator strengths of different 4f-4f bands and experimentally determined Judd-Ofelt intensity (T_{λ}) parameters are correlated with the binding of uracil. We also manifested particular interest in the effect of different solvent media on the relatively sharp absorption bands seen in the 400-620 nm regions.

II. MATERIALS AND METHODS

 $Pr(NO_3)_3$. $6H_2O$ (99% purity) from CDH, Mumbai, Zn(II) nitrate (Aldrich USA) and uracil of G R grade from SRL India, were used without further purification. The solvents used were CH₃CN, CH₃OH, DMF and Dioxane of A.R. grade from E.Merk.

The absorption spectral data are recorded on Perkin Elmer Lambda -35 UV-Vis Spectrometer equipped with a device for kinetic and high resolution spectral analysis. The concentrations of Pr(III) and Uracil and Zn(II) were maintained at 0.01 mol L^{-1} and spectral analysis were carried out by using different solvents. The temperature of all observations was maintained by using Perkin Elmer PTP1 Peltier -temperature system.

III. THEORETICAL METHODS

Judd-Ofelt[12,13] theory predicts that the intensity of

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4f-4f transition arise principally from forced electric dipole mechanism. Thus the electric dipole oscillator strength (P) is expressed as the product of T_{λ} parameters and appropriate transition matrix element U^(λ) at frequency of transition J \rightarrow J'.

$$P_{cal} = \sum_{\lambda=2,4,6} T_{\lambda} \upsilon \left(f^{n} \psi_{J} \| U^{\lambda} \| f^{n} \psi_{J'} \right)^{2}$$
(1)

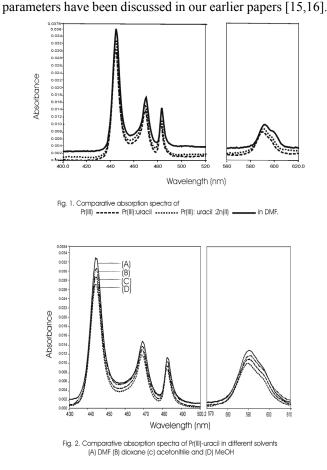
The Judd Ofelt intensity parameters are empirical, yet these show high sensitivity towards even minor changes in the coordination environment and symmetry of the molecule.[14]. These parameters have been widely used in the structure elucidation of lanthanoid coordination compounds in solution.

The energy of 4f-4f transitions comprises of two main components : coulombic (represented by inter-electronic repulsion F_k , Slater Condon) and spin-orbit (Lande- ξ_{4f}) components. The complexation of lanthanoids bring about lowering of nephelauxetic effect represented by nephelauxetic ratio ($\overline{\beta}$):

$$\beta_1 = \frac{F_k^c}{F_k^f} \quad \beta_2 = \frac{\xi_{4f}}{\xi_{4f}^f} \quad \overline{\beta} = \frac{\beta_1 + \beta_2}{2} \tag{2}$$

$$b^{\frac{1}{2}} = \begin{bmatrix} 1 - \beta \\ 2 \end{bmatrix}^{\frac{1}{2}} \qquad \delta = \begin{pmatrix} 1 - \beta \\ \beta \end{pmatrix} \times 100 \tag{3}$$

Bonding (b) and percent covalency parameter (δ) which are related to nephelauxetic ratio (β) are also used to describe the structural features of lanthanoid complexes. The significance and evaluation of these (Fk, $\overline{\beta}$, δ , Ek, ξ 4f and b)



IV. RESULTS AND DISCUSSIONS

From fig. 1, we can see that there is a red shift as uracil is added to Pr(III) and further shift towards longer wavelength is observed on addition of Zn(II). Table 1 shows the variation of the magnitude of energy interaction parameters like slator-Condon(F_k), spin-orbit coupling constant (ξ_{4F}), Racah parameters (E^{k}), nephelauxetic ratio (β), bonding parameter $(b^{1/2})$ and covalency parameter (δ) for Pr(III) and Pr(III):uracil and) in different solvents. The binding of Uracil to Pr(III) brings about changes in energies of various 4f-4f bands which causes the degree of lowering in the energy interaction parameters like Slator-Condon and inter-electronic repulsion parameters, which lead to nephelauxetic effect. The data in Table 1 shows that significant lowering of these parameters takes place in complexes as compared to the values for aquo ions indicating the expansion of the central metal ion orbital on complexation. Lowering of these parameters become more significant when Zn(II) is added to binary mixtures of Pr(III) and uracil (Table 3) . The positive values of $b^{1/2}$ indicates some covalent character in metal-ligand bond. The small value and small variation of it suggests that the 4f-orbitals are very slightly involved in the interaction of Uracil to Pr(III).

Since lanthanides are hard metal ions in the pearson classification scheme [17] they are preferentially attracted towards hard donor sites life oxygen atoms. This implies that interaction of Pr(III) with uracil is generally predominant with the carbonyl group of uracil. Hence the bonding between Pr(III) and uracil is basically electrostatic in nature as indicated by the absolute values of nephelauxetic effect (β), covalency (δ) and bonding parameters ($b^{1/2}$) thereby suggesting the mode of binding of uracil to Pr(III) involves predominantly ionic modes [18]. The absolute values of oscillator strengths and Judd-Ofelt intensity (T_{λ}) parameters are determined under different experimental conditions for the interaction Pr(III) and uracil (Table 2). This clearly shows a significant change in the oscillator strengths of 4f-4f bands, when Pr(III) interacts with uracil in the solution. Comparative absorption spectra of Pr(III); Pr(III)-uracil and Pr(III):uracil:Zn(II) in DMF (Fig. 1) clearly show that the addition of uracil to Pr(III) results in significant enhancement in the oscillator strengths of different 4f-4f transitions. We also observe noticeable increase in the magnitude of Judd-Ofelt parameters suggesting the binding of the uracil to Pr(III) in solution. This intensification of 4f-4f bands became more prominent when Zn(II) was added to the binary mixture of Pr(III) and uracil (Table 4). This clearly suggested that heterobimetallic interaction of uracil was much stronger than binary complexation of uracil. Therefore, we can say that different metal bindings are stimulative to each other.

 T_2 values appear to be negative which is meaningless. T_4 and T_6 are affected significantly. Both parameters are related to changes in symmetry properties of the complex species. At the same time, the extent of mixing of 4f and 4d orbital also influences T_6 parameter predominantly, while some influence on T_4 can also be extended. Comparative absorption spectra of Pr(III)-uracil in different aquated organic solvents (Fig. 2) clearly suggest the significant role of solvents on complexation. DMF appears to induce the strongest influence on Pr(III)-uracil complexes. The red shift is observed in the energies of all bands. The influence of solvents like methanol and acetonitrile are less on complexation of Pr(III)-uracil. Although we have found red shift in energies of all transitions, the effect is more pronounced in DMF. The red shift is due to the expansion of the metal orbital radius resulting in the decrease of inter-electronic repulsion which leads to the phenomenon of nephelauxetic effect and thus can lead to lowering of coordination number [19]. Table 2 also clearly shows that T_4 and T_6 parameters are effected significantly in the presence of different solvents, suggesting that not only immediate coordination environment of Pr(III), but symmetry of the complex species is also changed dramatically. These changes

are considered to be a good evidence for the involvement of uracil in the inner-sphere coordination of Pr(III). Though the ligand environment has a weak influence on the electronic cloud of the Pr(III), the 4f shell is efficiently shielded by the closed 5s and 5p shells. This clearly shows without doubt that the complexation of Pr(III) with uracil in presence and absence of Zn(II), is effected significantly by changes in the nature of solvent. All the results obtained clearly suggest that minor coordination changes in the Pr(III) complexes are caused by the different coordinating sites of uracil, coordination number, denticity, solvent nature and nature of Pr(III)-uracil bond, which do induce significant variation in the intensity of f-f transitions.

TABLE 1. CALCULATED VALUES OF ENERGY INTERACTION PARAMETERS (F_k), SPIN-ORBIT COUPLING CONSTANT (ξ_{4F}), RACAH PARAMETERS (E^k),
NEPHELAUXETIC RATIO (B), BONDINGPARAMETER ($b^{1/2}$) AND COVALENCY PARAMETER (Δ) FOR THE INTERACTION OF PR(III) WITH URACIL IN
DIFFERENT MEDIUM at 25°C

System										
	F_2	F_4	F ₆	ξ_{4f}	E^1	E^2	E^3	β	b ^{1/2}	δ
Pr(III) aquo ion	308.96	42.65	4.66	722.60	3508.10	23.73	614.22	0.9460	0.1632	5.6719
Pr(III):uracil										
МеОН										
DMF	308.95	42.65	4.66	722.24	3508.09	23.73	614.22	0.9463	0.1638	5.6719
	308.81	42.63	4.66	721.84	3506.48	23.72	613.94	0.9458	0.1646	5.7261
Acetonitrile	308.93	42.64	4.66	722.23	3507.88	23.73	614.18	0.9463	0.1639	5.6755
Dioxane	500.75	42.04	4.00	122.23	5507.00	23.15	014.10	0.9405	0.1057	5.0755
DMF:Dioxane	308.95	42.65	4.66	722.60	3508.10	23.74	614.22	0.9466	0.1634	5.6442
Divit Dioxale	308.83	42.63	4.66	722.14	3506.76	23.73	613.99	0.9461	0.1642	5.6991
DMF:Acetonitrile										
MeOH:Acetonitrile	308.62	42.60	4.66	720.69	3504.29	23.71	613.56	0.9448	0.1662	5.8454
	308.95	42.65	4.66	721.98	3508.07	23.73	614.22	0.9461	0.1641	5.6923
MeOH:Dioxane	209.02	12 (1	1.00	722 59	2507.96	22 72	(14.10	0.0465	0.1(25	5 (105
MeOH:DMF	308.93	42.64	4.66	722.58	3507.86	23.73	614.18	0.9465	0.1635	5.6495
	308.85	42.63	4.66	722.51	3506.92	23.73	614.02	0.9464	0.1638	5.6684

TABLE 2. CALCULATED VALUES OF OSCILLATOR STRENGTHS (P x 10⁶) AND JUDD-OFELT (TA x 10¹⁰) PARAMETERS FOR PR(III)-URACIL IN DIFFERENT MEDIUM.

	${}^{3}\text{H}_{4} \rightarrow {}^{3}\text{P}_{2}$		$^{3}\text{H}_{4} \rightarrow$	${}^{3}\text{H}_{4} \rightarrow {}^{3}\text{P}_{1}$		${}^{3}\text{H}_{4} \rightarrow {}^{3}\text{P}_{0}$		${}^{3}\text{H}_{4} \rightarrow {}^{1}\text{D}_{2}$			
System	Pobs	P _{cal}	Pobs	P _{cal}	Pobs	P _{cal}	Pobs	P _{cal}	T ₂	T_4	T_6
Pr(III) aquo ion	3.2475	3.2475	0.7819	0.4360	0.1517	0.4576	1.18 00	1.1800	-8.03	1.69	10.30
Pr(III):uracil MeOH											
							1.18		52.9		
DMF	3.2490	3.2490	0.7820	0.4730	0.1615	0.4657	01 1.15	1.1801	7 -27.6	1.30	10.31
Acetonitrile	3.5459	3.5459	0.8785	0.6903	0.4943	0.6796	66 0.94	1.1566	9 -18.1	1.89	11.12
Dioxane	3.5175	3.5175	0.8211	0.6319	0.4359	0.6222	67 1.06	0.9467	0	1.73	11.07
DMF:Dioxane	3.5283	3.5283	0.9101	0.6955	0.4735	0.6847	7 0.94	1.067	-8.16	1.91	11.06
	3.1170	3.1170	0.8307	0.6346	0.4315	0.6246	7	0.947	-8.34	1.74	9.76
DMF:Acetonitrile	3.2677	3.2677	0.8426	0.6515	0.4532	0.6414	0.98 25	0.9825	-6.19	1.79	10.24
MeOH:Acetonitri le MeOH:Dioxane	2.9153	2.9153	0.9365	0.6966	0.4497	0.6859	0.85 25 0.93	0.8525	-0.11	1.91	9.05
MeOH:DIoxane	3.2785	3.2785	0.6946	0.5536	0.4062	0.545	0.93 6 0.75	0.936	-4.62	1.52	10.34
MeOn.DMF	1.2912	1.2912	0.9081	0.6939	0.4721	0.6830	0.75 40	0.7540	-84.0	1.90	3.72

ystem										
,~	F ₂	F₄	F 6 2	- 4f	E^1	E ²	E ³ f	b b ¹	/2	5
МеОН	308.95	42.65	4.66	722.28	3508.25	23.74	614.25	0.9464	0.1637	5.6663
DMF	308.72	42.61	4.66	722.14	3505.49	23.72	613.77	0.9461	0.1644	5.7169
Acetonitrile	308.91	42.63	4.66	722.19	3508.18	23.74	614.14	0.9463	0.1639	5.6743
Dioxane	308.91	42.63	4.66	722.28	3507.80	23.73	614.17	0.9463	0.1638	5.6731
DMF:Dioxane	308.75	42.62	4.66	721.96	3505.77	23.72	613.81	0.9468	0.1646	5.7267
DMF:Acetonitrile	308.75	42.62	4.66	721.96	3505.77	23.72	613.81	0.9468	0.1646	5.7267
MeOH:Acetonitrile	308.87	42.64	4.66	722.08	3507.18	23.73	614.06	0.9465	0.1642	5.6970
MeOH:Dioxane	308.94	42.64	4.66	722.31	3507.90	23.73	614.19	0.9466	0.1638	5.6695
MeOH:DMF	308.76	42.61	4.66	722.25	3505.95	23.72	613.85	0.9467	0.1642	5.7020

TABLE 3. CALCULATED VALUES OF ENERGY INTERACTION PARAMETERS (F_k), SPIN-ORBIT COUPLING CONSTANT (ξ_{4F}), RACAH PARAMETERS (E^k),NEPHELAUXETIC RATIO (B), BONDING PARAMETER ($b^{1/2}$) AND COVALENCY PARAMETER (Δ) for the Interaction of Pr(III) with Uracil in Presence of Zn(II) in Different Medium at 25°C.

TABLE 4. CALCULATED VALUES OF OSCILLATOR STRENGTHS (P x 10^6) and Judd-Ofelt ($T_\lambda x 10^{10}$) Parameters for Pr(III):uracil:Zn(II) in Different Medium.

System	$^{3}\mathrm{H}_{4} \rightarrow$	$^{3}P_{2}$	$^{3}\text{H}_{4} \rightarrow$	${}^{3}P_{1}$	$^{3}H_{4} \rightarrow$	${}^{3}P_{0}$	$^{3}\text{H}_{4} \rightarrow$	$^{1}D_{2}$	T_2	T_4	T_6
	P _{obs}	\mathbf{P}_{cal}	P_{obs}	\mathbf{P}_{cal}	\mathbf{P}_{obs}	P _{cal}	P _{obs}	P _{cal}	-		
MeOH											
	3.4030	3.4030	0.8383	0.6529	0.4603	0.6428	1.1801	1.1181	-27.77	1.79	10.6
DMF	3.7927	3.7927	1.8661	3.7665	0.4596	0.6561	1.1901	1.1901	-22.15	1.83	10.9
Acetonitrile											
	3.5311	3.5311	0.726	0.7731	0.4137	0.5642	0.9252	0.9252	-10.53	1.57	11.5
Dioxane	3.4582	3.4582	0.4665	0.6556	0.4378	0.6486	0.9949	0.9949	-21.63	1.25	11.0
DMF:Dioxane	3.3257	3.2257	0.8679	0.6801	0.4845	0.6694	1.1828	1.1828	-14.61	1.86	05.4
DMF:Acetonitrile	3.3225	3.3225	0.8716	0.6714	0.4639	0.661	1.0594	1.0594	-20.16	1.84	10.4
MeOH:Acetonitrile	3.2098	3.2098	0.8278	0.6971	0.4396	0.6273	0.9422	0.9422	-18.12	1.75	10.0
MeOH:Dioxane	3.4869	3.4869	0.8151	0.6385	0.4548	0.6287	1.0323	-1.0323	-23.35	1.75	10.9
MeOH:DMF	4.6300	4.630	0.9564	0.7264	0.4886	0.715	1.2222	1.2222	-28.85	1.99	14.6

V. CONCLUSIONS

From the above discussion through variation in the magnitude of energy interaction and intensity parameters we can suggest that the heterobimetallic complexation of uracil is more stable than binary mixtures of Pr(III) and uracil. Their kinetic study on simultaneous coordination of uracil with Pr(III) and Zn(II) using 4f-4f transition spectra is going on for next paper.

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