SPECTRAL INVESTIGATION AND CALCULATION OF BONDING PARAMETERS FOR Nd (III) SYSTEM WITH 2-AMINOBENZOTHIAZOLE DERIVATIVES AND CODOPING OF Ca^{+2}, Mg^{+2}, Sr^{+2} METAL IONS

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ABSTRACT: -
Electronic absorption studies of the complexes with 2-amino benzothiazole derivatives doped in Nd (III) ion have been calculated. In present work, we describes the spectral characterization of the complexes of benzothiazole derivatives with Nd(III) ion in agreement to Green chemistry approach. For eleven different peaks, viz $^4I_{9/2}\rightarrow{}^2P_{1/2}$, $^4I_{9/2}\rightarrow{}^4G_{11/2}$,$^4I_{9/2}\rightarrow{}^2G_{9/2}$,$^4I_{9/2}\rightarrow{}^4G_{9/2}$,$^4I_{9/2}\rightarrow{}^4G_{7/2}$, $^4I_{9/2}\rightarrow{}^2G_{7/2}$,$^4I_{9/2}\rightarrow{}^4F_{9/2}$, $^4I_{9/2}\rightarrow{}^4S_{3/2}$, $^4I_{9/2}\rightarrow{}^4F_{7/2}$, $^4I_{9/2}\rightarrow{}^4F_{5/2}$, and $^4I_{9/2}\rightarrow{}^4F_{3/2}$ transitions of Nd(III) have been observed . Various spectroscopic parameters viz- Interaction Parameters viz: Slater- Condon F_2, F_4,& F_6 and Lande parameter or Spin-Orbit Coupling Constant (ζ_{4f}, Racah Parameters or Energy parameters (E^1, E^2 and E^3):Bonding parameters : Nephelauxetic ratio (β), Bonding($b^{1/2}$), Sinha Covalency (δ%) and Covalency-Angular Overlap (η) have been computed using partial and multiple regression methods and evaluated by expressing energy as Taylor series. This work describes investigations on Nd (III)-2-amino benzothiazole system. Effect of codoped viz. Ca^{+2},Mg^{+2},Sr^{+2} metal ions have been analyzed and it has been observed that degree of covalency have been raised in presence of Ca^{+2},Mg^{+2},Sr^{+2} metal ions.

KEYWORDS-Spectral characterization, Interaction Parameters, Bonding Parameters, Nd(III) Ion and benzothiazole derivative

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INTRODUCTION-

Lanthanide chemistry has attracted much attention in last decade. Lanthanides are used as structural and functional probes to understand bimolecular structures, conformations and properties [1–3]. The construction and characterizations of lanthanide complexes are currently of great interest because of their unique physico-chemical properties and various applications as laser material. Various concepts and hypotheses have been modified or rejected as the science of coordination chemistry progresses. In coordination chemistry, Complex formation processes are a key factor in a number of research area like analytical, bioinorganic, clinical and biochemical aspects [4-7].

Under certain experimental and physiological conditions. Lanthanide (III) ions are hard metal ions, favour hard donor ligands such as oxygen, nitrogen and halogen. In multidentate large biological structures, there are many ligands containing oxygen, nitrogen, sulphur, halogen and phosphorous donor atoms in the form of functional groups. In complexation, lanthanides prefer donor atoms in the following order: O>N>S and F>Cl. This indicates that the lanthanide ion has a strong preference for 'O' and 'N' donor atoms [8-11]. In this work, we discuss coordination chemistry of Nd(III) ion with benzothiazole derivatives having N-donar sites. benzothiazole derivatives have attracted significant interest in recent time for their usefulness in synthetic heterocyclic chemistry, analytical chemistry and pharmacology.

In the present electronic absorption study, energy interaction parameters and covalent bonding parameter of Nd(III) ion with 2-amino benzo thiazole derivatives in alcoholic :water(1:1) medium have been evaluated using Judd Ofelt theory. The intensities of the solution spectra of most trivalent lanthanides have been interpreted by means of Judd-ofelt theory. The $f \leftrightarrow f$ transitions are assumed to be forced electric dipole in nature, occurring by the mixing of a higher energy configurations of opposite parity with $4f^N$ configuration via the odd term in crystal field expansion.

MATERIAL AND METHODS:

Nd (III) carbonate hydrate, alfa was purchased from M/s Sita Trading Company and ligand were perchased from Sharda Enterprises. The solvents used in doped system are Ethyl Alcohol (AR 99.9%, Jiangsu-Hliaxi International Trade Co. Ltd., Made in China).In the present study Nd (III) ions have been doped and codoped Ca^{+2},Mg^{+2},$Sr^{+2}$with the saturated solution of ligands in Alc: Water(1:1) medium. The saturated solutions have been prepared by dissolving requisite amount of ligand and metal. The saturated solution of ligand and metal Nd(III) ion of 0.1 M were prepared is used for experiment. The solution spectra of each system at room temperature in 1:3 ratio (Metal: Ligand) has been recorded by using standard UV-VIS spectrophotometer 3092. Slater Condon F_2, F_4,& F_6 have been computed by using theses spectral data. In the present study measurement was done in the range of 400-900 nm regions.

CALCULATION OF PARAMETERS-

1. Slater- condon- Lande Equation: The Slater- Condon F_2, F_4,& F_6 and Lande parameter or Spin-Orbit Coupling Constant (ζ_{4f}) may be evaluated by expressing energy as Taylor series expansion may be used to express the energy of jth electronic energy level . In the first order approximation the energy E_j of a J-level is given by the following equation-

$$E_j(F_k , \zeta_{4f}) = E_{oj}(F^o_k , \zeta^o_{4f}) + \sum (\partial E / \partial F_k)\Delta F_k + (\partial E_j / \partial \zeta_{4f})\Delta\zeta_{4f} \quad k = 2, 4, 6 \text{ ---------- (5)}$$

$$E_{obs} = E_{oj} + (\partial E_j/\partial F_2)\,\Delta F_2 + (\partial E_j/\partial F_4)\,\Delta F_4 + (\partial E_j/\partial F_6)\,\Delta F_6 + (\partial E_j/\partial\zeta_{4f})\,\Delta\zeta_{4f} \text{ ----------(6)}$$

$$E_{obs} - E_{oj} = (\partial E_j/\partial F_2)\,\Delta F_2 + (\partial E_j/\partial F_4)\,\Delta F_4 + (\partial E_j/\partial F_6)\,\Delta F_6 + (\partial E_j/\partial\zeta_{4f})\,\Delta\zeta_{4f} \text{ ----------(7)}$$

$$[(E_{obs} - E_{oj}) \div (\partial E_j/\partial F_2)] = \Delta F_2 + [(\partial E_j/\partial F_4) \div (\partial E_j/\partial F_2)]\,\Delta F_4 + [(\partial E_j/\partial F_6) \div (\partial E_j/\partial F_2)]\,\Delta F_6 + [(\partial E_j/\partial\zeta_{4f}) \div (\partial E_j/\partial F_2)]\,\Delta\zeta_{4f} \text{ ----------(8)}$$

Where, E_j = Energy of jth level

E_{oj} = Zero-order Energy of jth level

ΔF_k = Small changes in the Slater-Condon parameters

$\Delta\zeta_{4f}$ = Small changes in the Lande parameters

$(\partial E_j / \partial F_k)$ = Partial derivatives with respect to Fk

$(\partial E_j / \partial \zeta_{4f})$ = Partial derivatives with respect to ζ_{4f}

The values of ΔF_k and $\Delta\zeta_{4f}$ may be evaluated using observed energy values (E_j), reported values of zero-order energies (E_{oj}) and partial derivatives $(\partial E_j / \partial F_k)$ and $(\partial E_j / \partial \zeta_{4f})$ by partial and multiple regression method (12-15).

The values of Slater-Condon parameters (F_k) and Lande para-meters (ζ_{4f}) are then evaluated using following equations-

$$F_k = F^o_k + \Delta F_k \quad (9)$$

$$\zeta_{4f} = \zeta^o_{4f} + \Delta\zeta_{4f} \quad (10)$$

Where, $\Delta Fk << F^o_k$, $\Delta\zeta_{4f} << \zeta^o_{4f}$ and F^o_k and ζ^o_{4f} are the zero -order values of Slater-Condon and spin-orbit interaction parameters as reported by Wong (16).

Racah Parameters or Energy parameters (E^1, E^2 and E^3):

The Racah parameters E^k (k =1,2, and 3) are related to F_k (k=2,4and6) parameters by the following relations-

$$E^1 = (1/9)\,(70F_2 + 231F_4 + 2002F_6) \text{ ----------} (11)$$

$$E^2 = (1/9)\,(F_2 - 3F_4 + 7F_6) \text{ .....} (12)$$

$$E^3 = (1/3)\,(5F_2 + 6F_4 - 91F_6) \text{----------} (13)$$

Or

By assuming wave functions to be hydrogenic as E^K's (racah parameters) are the linear combination of F_K's, they may be calculated by using the following relation the Racah parameters reduces to in case of Pr^{+3}complexes which have been evaluated using these equations.

$$E^1 = 14.6818F_2 \text{.....} (11)$$

$$E^2 = 0.0768F_2 \text{.....} (12)$$

$$E^3 = 1.4844\,F_2 \text{.....} (13)$$

(c) R.M.S. Deviations (Energy):

The energies Ej of the electronic sates have been estimated by using the computed values of the parameters. For comparison between calculated (Ecal) and observed (E_{obs}) values of energy, the value of r.m.s. deviations (σ) have been computed by using the relations-

$$\sigma = \{\Sigma\,(E_{cal} - E_{obs})\,/N\}1/2 \text{.....} (14)$$

Where, N =11, Number of energy level fitted

2. Bonding Parameters:

These parameters consist of Nephelauxetic ratio (β), bonding parameter ($b^{1/2}$), Sinha's Covalency Parameters (δ%) and Covalency angular overlap Parameter (η) (17-25).

Nephelauxetic Ratio (β)

The effect of complexation on the free ion is that it shows red shift in the electronic transition. This red shift of the bands is due to the expansion of the orbital radius of central metal ion, resulting the lowering of the interelectronic repulsion parameters. This phenomena is called Nephelauxetic effect. This effect can be defined in terms of Nephelauxetic ratio (β) as follows

$\beta = \nu_c/\nu_f$ .....(**VIII**)

Where, ν_c and ν_f are wave numbers of f-f band for spectra of complex and free ion respectively. This relation reflects how effectively bands of metal ions are shifted in presence of ligand environment. It has been generally agreed that ligands cause slight red shifts in bands of free metal ions.

Bonding parameter ($b^{1/2}$)

The Nephelauxetic ratio value can be used to calculate the bonding parameter, from the relation given as –

$$\mathbf{b^{1/2}} = [1/2\,(1-(\beta)]^{1/2} \text{ .....} (\mathbf{IX})$$

Positive value of bonding parameter ($b^{1/2}$) indicates covalent character in metal-ligand interaction while negative value depicts ionic character in metal-ligand interaction [16].

(c) Sinha's Covalency Parameters (δ %)

Percentage covalent character in metal ligand bond can be expressed by Sinha's covalency parameter (δ %),

$$\delta = [(1-\beta)/\beta\,]\,X\,100 \text{.....} (\mathbf{X})$$

(d) Covalency angular overlap parameter (η)

Covalency angular overlap parameter (η) represents magnitude of overlapping between

Ligand and metal ion orbital.

$$\eta = [(1 - \beta^{1/2})/ \beta] \text{ ..... (XI)}$$

High and Low value of bonding parameters represent strong and weak covalent character in metal-ligand bond respectively.

RESULTS AND DISCUSSION-

Covalency parameters, viz: Bonding Parameter ($b^{1/2}$), Nephelauxetic ratio (β), Sinha's covalency parameter δ and covalency angular overlap parameter (η) are given in Table I-V). The value of Nephelauxetic ratio (β) has been found to be less than one and Bonding parameter ($b^{1/2}$), has been observed positive for all metal-ligand systems indicates covalency between Nd(III) ion and 2-amino benzothiazole derivatives moiety. Lower value of β showing a red shift in electronic absorption spectra which causes absorption bands to shift towards higher wavelengths or lower wave numbers. Sinha's covalency parameter (δ %) and Covalency angular overlap parameter (η) also prefers the covalent characters in M-L bond due to decrease in inter electronic repulsion parameter, highest red shift is observed in Nd(III) ion and 2-amino benzothiazole derivatives during codoping of Ca^{+2},Mg^{+2},Sr^{+2} metal ions. Higher value of red shift shows higher extent of mixing of 4f orbital and value of $b^{1/2}$, Sinha's covalency factor δ and covalency angular overlap parameter η represent covalent bonding between Nd(III) ion and 2-amino benzothiazole derivatives.

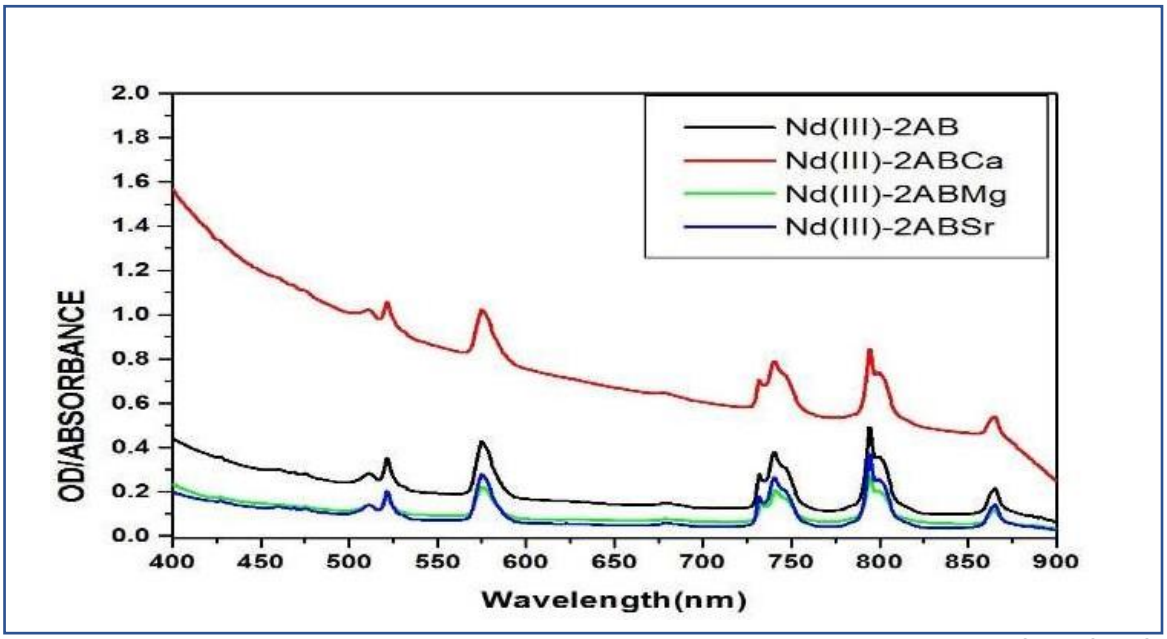

FIG.1- Variation of optical density with wavelength for Nd(III)-2AB and codoping of Ca^{+2},Mg^{+2},Sr^{+2} metal ion in Alc: Water(1:1) medium

TABLE. I

Computed value of energy interaction: Slater-Condon $F_k(cm^{-1})$,Spin-Orbit interaction Lande ζ_{4f} (cm^{-1}) Racah E^k (cm^{-1}) perameters and Hydrogenic Ratio (F_4/F_2) , (F_6/F_2) and (E^1/E^3) , (E^2/E^3) of Nd (III)-2AB Complexe and Codoping of Ca^{+2},Mg^{+2},Sr^{+2} ions in Alc: Water(1:1) medium

Complex		Nd (III)-2AB	Nd (III)-2AB Ca^{+2}	Nd (III)-2AB Mg^{+2}	Nd (III)-2AB Sr^{+2}
S.N.	Parameters				
1	E_1	5157.8181	5141.951988	5131.42416	5137.543867
2	E_2	26.685398	26.67216477	26.5381768	26.48425854
3	E_3	507.70004	507.5137738	508.4062215	508.8516065
4	F_2	346.63168	346.1600729	345.6869495	345.7657264
5	F_4	48.305411	48.11202161	48.24787093	48.44151537
6	F_6	5.4933047	5.460782118	5.414322073	5.416735216
7	ζ_{4f}	811.61004	816.3610168	813.2002385	807.9925112
8	$\%r\zeta_{4f}$	6.5658287	6.018906387	6.382768062	6.982269887
9	$\%rF_2$	-0.662166	-0.52521127	-0.387815798	-0.410692684
10	F_4/F_2	0.1393566	0.138987784	0.139570993	0.14009924
11	F_6/F_2	0.0158477	0.015775309	0.015662501	0.015665911
12	E_1/E_3	10.159184	10.13165012	10.09315768	10.0963499
13	E_2/E_3	0.0525613	0.052554563	0.052198765	0.052047116

TABLE. II Computed values of Bonding parameters: Nephelauxetic ratio (β), Bonding ($b^{1/2}$), Sinha Covalency (δ %) and Covalency-Angular Overlap (η) Parameters of Nd (III)2AB Complex in Alc: Water(1:1) medium

sn.	Transition	λc(nm)	Vc (cm^{-1})	(β)	($b^{1/2}$)	δ%	(η)
1	$^4I_{9/2}\rightarrow{}^2P_{1/2}$	426	23474.1784	1.0023474	0.03425944	-0.234192	-0.00117
2	$^4I_{9/2}\rightarrow{}^4G_{11/2}$	460	21739.1304	1.0021739	0.03296902	-0.2169197	-0.00109
3	$^4I_{9/2}\rightarrow{}^2G_{9/2}$	476	21008.4034	0.9978992	0.03241019	0.21052632	0.001052
4	$^4I_{9/2}\rightarrow{}^4G_{9/2}$	512.5	19512.1951	0.9990244	0.02208631	0.09765625	0.000488
5	$^4I_{9/2}\rightarrow{}^4G_{7/2}$	521.5	19175.4554	0.9990412	0.02189489	0.09596929	0.00048
6	$^4I_{9/2}\rightarrow{}^2G_{7/2}$	574.5	17406.4404	1.0008703	0.02086051	-0.0869565	-0.00043
7	$^4I_{9/2}\rightarrow{}^4F_{9/2}$	679.5	14716.7035	0.9992642	0.01918118	0.0736377	0.000368
8	$^4I_{9/2}\rightarrow{}^4S_{3/2}$	732.5	13651.8771	0.9993174	0.01847422	0.06830601	0.000341
9	$^4I_{9/2}\rightarrow{}^4F_{7/2}$	739.5	13522.6504	1.0006761	0.01838658	-0.0675676	-0.00034
10	$^4I_{9/2}\rightarrow{}^4F_{5/2}$	793.5	12602.3945	1.0006301	0.01774993	-0.0629723	-0.00031
11	$^4I_{9/2}\rightarrow{}^4F_{3/2}$	864.5	11567.38	1.0005784	0.01700543	-0.0578035	-0.00029

TABLE. III: Computed values of bonding parameters: Nephelauxetic ratio (β), Bonding ($b^{1/2}$), Sinha Covalency (δ %) and Covalency-Angular Overlap (η) Parameters of Nd (III)2AB Complex with Codoping of Ca^{+2} ion in Alc: Water(1:1) medium

sn.	Transition	λc(nm)	Vc (cm^{-1})	(β)	($b^{1/2}$)	δ%	(η)
1	$^4I_{9/2}\rightarrow{}^2P_{1/2}$	426	23474.1784	-	-	-	-
2	$^4I_{9/2}\rightarrow{}^4G_{11/2}$	460	21739.1304	-	-	-	-
3	$^4I_{9/2}\rightarrow{}^2G_{9/2}$	476	21008.4034	0.9978992	0.03241019	0.21052632	0.001052
4	$^4I_{9/2}\rightarrow{}^4G_{9/2}$	510	19607.8431	1.0039216	0.04428074	-0.390625	-0.00196
5	$^4I_{9/2}\rightarrow{}^4G_{7/2}$	521	19193.858	1	0	0	0
6	$^4I_{9/2}\rightarrow{}^2G_{7/2}$	575.5	17376.1946	0.9991312	0.02084238	0.08695652	0.000435
7	$^4I_{9/2}\rightarrow{}^4F_{9/2}$	679	14727.5405	1	0	0	0
8	$^4I_{9/2}\rightarrow{}^4S_{3/2}$	732.5	13651.8771	0.9993174	0.01847422	0.06830601	0.000341
9	$^4I_{9/2}\rightarrow{}^4F_{7/2}$	739	13531.7997	1.0013532	0.02601134	-0.1351351	-0.00068
10	$^4I_{9/2}\rightarrow{}^4F_{5/2}$	794.5	12586.5324	0.9993707	0.01773875	0.06297229	0.000315
11	$^4I_{9/2}\rightarrow{}^4F_{3/2}$	864	11574.0741	1.0011574	0.02405626	-0.1156069	-0.00058

TABLE. IV: Computed values of Bonding parameters: Nephelauxetic ratio (β), Bonding ($b^{1/2}$), Sinha Covalency (δ %) and Covalency-Angular Overlap (η) Parameters of Nd (III)2AB Complex with Codoping of Mg^{+2} ion in Alc: Water(1:1) medium

sn.	Transition	λc(nm)	Vc (cm^{-1})	(β)	($b^{1/2}$)	δ%	(η)
1	$^4I_{9/2}\rightarrow{}^2P_{1/2}$	426	23474.1784	-	-	-	-
2	$^4I_{9/2}\rightarrow{}^4G_{11/2}$	460	21739.1304	-	-	-	-
3	$^4I_{9/2}\rightarrow{}^2G_{9/2}$	476	21008.4034	-	-	-	-
4	$^4I_{9/2}\rightarrow{}^4G_{9/2}$	509	19646.365	1.00589391	0.05428586	-0.58594	-0.002934
5	$^4I_{9/2}\rightarrow{}^4G_{7/2}$	520.5	19212.296	1.00096061	0.02191592	-0.09597	-0.00048
6	$^4I_{9/2}\rightarrow{}^2G_{7/2}$	575	17391.304	1	0	0	0
7	$^4I_{9/2}\rightarrow{}^4F_{9/2}$	678.5	14738.394	1.00073692	0.01919531	-0.07364	-0.0003683
8	$^4I_{9/2}\rightarrow{}^4S_{3/2}$	732	13661.202	1	0	0	0
9	$^4I_{9/2}\rightarrow{}^4F_{7/2}$	738.5	13540.961	1.00203114	0.03186804	-0.2027	-0.001014
10	$^4I_{9/2}\rightarrow{}^4F_{5/2}$	794	12594.458	1	0	0	0
11	$^4I_{9/2}\rightarrow{}^4F_{3/2}$	864.5	11567.38	1.00057837	0.01700543	-0.0578	-0.0002891

TABLE. V Computed values of Bonding parameters: Nephelauxetic ratio (β), Bonding ($b^{1/2}$), Sinha Covalency (δ %) and Covalency-Angular Overlap (η) Parameters of Nd (III) 2AB Complex with Codoping of Sr^{+2} ion in Alc: Water(1:1) medium

sn.	Transition	λc(nm)	Vc (cm^{-1})	(β)	($b^{1/2}$)	δ%	(η)
1	$^4I_{9/2}\rightarrow{}^2P_{1/2}$	426	23474.1784	-	-	-	-
2	$^4I_{9/2}\rightarrow{}^4G_{11/2}$	460	21739.1304	-	-	-	-
3	$^4I_{9/2}\rightarrow{}^2G_{9/2}$	476	21008.4034	-	-	-	-
4	$^4I_{9/2}\rightarrow{}^4G_{9/2}$	511	19569.472	1.00195695	0.03128056	-0.19531	-0.000977
5	$^4I_{9/2}\rightarrow{}^4G_{7/2}$	520	19230.769	1.00192308	0.03100868	-0.19194	-0.0009602
6	$^4I_{9/2}\rightarrow{}^2G_{7/2}$	574.5	17406.44	1.00087032	0.02086051	-0.08696	-0.0004349
7	$^4I_{9/2}\rightarrow{}^4F_{9/2}$	679.5	14716.703	0.99926416	0.01918118	0.073638	0.00036812
8	$^4I_{9/2}\rightarrow{}^4S_{3/2}$	731.5	13670.54	1.00068353	0.01848685	-0.06831	-0.0003416
9	$^4I_{9/2}\rightarrow{}^4F_{7/2}$	739	13531.8	1.00135318	0.02601134	-0.13514	-0.0006759
10	$^4I_{9/2}\rightarrow{}^4F_{5/2}$	793	12610.34	1.00126103	0.0251101	-0.12594	-0.0006299
11	$^4I_{9/2}\rightarrow{}^4F_{3/2}$	864	11574.074	1.00115741	0.02405626	-0.11561	-0.0005782

CONCLUSION:

The results reported in this work indicated that Nd (III) Complexes with 2-amino benzo thiazole derivatives with nitrogen,sulpher donor ligand creates greater extent of intensification to transitions in Alc: Water(1:1) medium. A greater extent of bonding between doped systems of Nd^{+3} ion with 2-amino benzo thiazole derivatives is observed ,which provides better ways to species for interaction. A distinct increase in intensity have been observed for all Nd (III)-2AB codoped of Ca^{+2},Mg^{+2},Sr^{+2} metalmetal ions systems . The value of nephelauxetic ratio (β), Sinha's covalency parameter (δ %), bonding parameter ($b^{1/2}$) and covalency angular parameter indicate the existence of covalent character in metal-ligand linkage in Nd (III)-2AB and codoped Ca^{+2},Mg^{+2},Sr^{+2} metalion alcoholic: water (1:1) medium. In case of codoping of Ca^{+2},Mg^{+2},Sr^{+2} metal ions, transitions like $^4I_{9/2}\rightarrow{}^2P_{1/2}$, $^4I_{9/2}\rightarrow{}^4G_{11/2}$,$^4I_{9/2}\rightarrow{}^2G_{9/2}$ disappeared.

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