SPECTROPHOTOMETRIC/SPECTROFLUORIMETRIC DETERMINATION OF RARE EARTH MIXTURES BY THEIR CHARACTERISTIC ABSORPTION OR FLUORESCENCE OF 4f ELECTRON TRANSITIONS

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Abstract

This review deals with the spectrophotometric and/or spectrofluorimetric determination of individual rare earth elements in rare earth mixtures by their characteristic absorption or fluorescence of 4f electron transitions, including conditions of maximal absorption or fluorescence, molar absorptivity, ranges of linearity, detection limits, interference, and application of these methods with 136 references cited.

Spectrophotometry

Lately, the awareness of the good characteristics of rare earth elements (RE) has increased [1]. About the spectrophotometric analysis of rare earths, there have been many reviews [2-5]. In addition, the rare earth (III) perchlorates, nitrates and chlorides except for La, Y and Lu have sharp narrow characteristic absorption peaks for the 4f electron transitions between 270 nm and 1200 nm. By making use of these characteristic peaks, individual rare earth elements are determined directly without any chemical separation. Recently, owing to the development of new model spectrophotometers and the discovery of various complexing agent/surfactant systems, the sensitivity and selectivity in the determination of some individual rare earth elements in their mixtures have become much higher. Their molar absorptivities are increased from 2.5-12 to 1400-4768 [6, 29]. These methods have been used for the determination of individual rare earths in mixed rare earth material. Spectrophotometric procedures based on the characteristic peaks are summarized, ordered according to various classes of reagents.

Amino-carboxyl complexing agents:

It has been reported that the characteristic peaks of Nd at 520 nm and 575 nm are split into four and five sharp peaks, and the sensitivity increases as EDTA or pH increases in the RE - EDTA systems [7-9]. In the pH range 2.5-10.5, the tetraethylenepentamineheptaacetic acid can form 1:1 complexes with rare earths [10], where the amplitude of the absorption bands of 4f electron transitions for Pr, Nd, Sm, Eu, Gd, Dy and Er are greatly enhanced. Their sensitivities are increased by a factor of about 50 (in Pr, 25), compared with those of the chlorides. The stability of the complexes is increased with increasing atomic number. Ordinary anions and cations cause no interference, but Sm and Eu interfere with each other in their determination. In 40 per cent hexamine solution, rare earths form ternary complexes with EDTA and $H_2C_8O_4$ [11-13], which can enhance the amplitude of the characteristic absorption peaks for Pr, Nd, Sm, Eu, Ho and Er. The sensitivities of their third-order derivative spectra are increased by a factor of

2-11. For the determination of Sm and Eu in rare earth mixtures, the relative error is less than 1 per cent. The triethylenetetraamine-hexaacetic acid (TTHA) can also increase the characteristic absorption of rare earth elements [14], and the sensitivities are increased even more than those with EDTA or DTPA. For the direct determination of Pr and Nd in rare earth mixtures, the relative standard deviation is less than 1.9 per cent.

β-Diketones:

In water-acetone medium, dibenzolmethane (DBM) can form stable complexes with rare earth elements [15, 16]. The absorption peaks of Pr, Nd, Eu, Ho, Er and Tm in the visible region are enhanced, the absorption at λmax being increased by factors of 2.2, 9, 3, 3.4, 17 and 4, respectively, compared with those of the chlorides. The sensitivities for Ho, Er and Tm are 1.3μg/ml, 3.7μg/ml and 2.1μg/ml. In the presence of pyridine (Py), α-aminepyridine (αApy) or diphenylguanidine (DPG), rare earths can form extractable ternary complexes with dibenzolmethane (DBM) or thenoyltrifluoroacetone (TTA). For the RE-TTA-Py system, the sensitivities for Nd, Tm, Er, Ho and Eu are 6.7, 8.0, 20, 30 and 6.3 times higher than those of the chlorides. The relative error is less than 4 per cent for the determination of an individual rare earth in a mixture of lanthanides [17]. In acetic acid (pH>2.5), rare earths can form two extractable ternary complexes, $RE(TTA)_3$.TOPO and $RE(TTA)_3$.2TOPO, with TTA and Tri-n-octyl-phosphine oxide (TOPO), which can enhance the characteristic peaks of Nd, Pr, Eu, Ho, Er and Tm. The sensitivities for Nd, Er and Ho are increased by a factor of 7, 22 and 26 respectively. In the concentration range 0-1.5 mg/ml of Nd, Ho and Er can obey Beer's Law, where Ce causes serious interference for the determination of Er and Ho, Sm, Tb, Yb and Dy have very weak absorption, and other lanthanides have no absorption; the method is therefore used to determine Nd, Ho and Er directly in the presence of other lanthanides (except for Ce) [18]. Rare earth elements can form 1:3:2 complexes with 1-phenyl-3-methyl-4-benzoyl-pyrazol-5-one (PMBP) and 8-hydroxyquinoline (HQ) [19], where the molar absorptivities obtained for Nd, Ho and Er are 20, 65 and 20 $l.mol^{-1}.cm^{-1}$ respectively, and 1:1:1

complexes with acetylacetone (AA) and citric acid [20], where the molar absorptivities evaluated for Nd, Ho and Er are 10.3, 18.5 and 10.0 $1.mol^{-1}.cm^{-1}$ respectively. In ethanol medium, rare earths can form 1:4:8 complexes with PMBP and DPG [21], their fourth-order derivative molar absorptivities obtained for Nd, Ho and Er are 123, 20 and 30 $1.mol^{-1}.cm^{-1}.nm^{-4}$ respectively, and the concentration range obeying Beer's Law is 0-1.0mg/ml. Rare earths can also form 1:1:1 complexes with 1-phenyl-3-methyl-4-heptafluorobutyryl-5-pyrazolone and diantipyrylmethane [22], where the molar absorptivities calculated for Nd, Ho and Er are 23.4, 47.6 and 25.6 $1.mol^{-1}.cm^{-1}$, respectively; the structure of the complexes is discussed and the oscillator strengths of 4f electron transitions are also calculated. Chen et al. [23] reported that RE(Nd, Ho, Er) can form 1:4:1 complexes with 1-phenyl-3-methyl-4-dichloroacetylpyrazolone-5 and quinoline, their characteristic peaks are enhanced and the shapes of the peaks show obvious changes; the molar absorptivities obtained for Nd, Ho and Er are 17.4, 50.7 and 19.7 $1.mol^{-1}.cm^{-1}$, respectively. Pr(0.5μg), Sm(0.05mg), Eu(0.76mg), Ce(0.42mg) and Dy(5.2μg) cause no interference in the determination of 0.3mg/ml Nd; and La(20μg), Sm(15μg), Eu(9.1μg), Ce(5.6μg) and Dy(5.1μg) produce no interference but Pr(5μg) cause interference in the determination of Er. In studies on RE-TTA-OB systems (OB represents organic bases), Zhao et al. [24] discovered that when organic bases are pyridine, 4-methylpyridine, trimethylpyridine and 2,6-dimethylpyridine, respectively, the sensitivities for the presence of 2,6-dimethylpyridine is highest, as a result of its alkalinity being stronger than that of trimethylpyridine. Investigation of the RE-TTA-2,6-dimethylpyridine system shows that in 5 ml solution, Ho exceeding 0.2mg causes interference in the determination of 0.5mg Pr; and Pr and Ho which exceed 0.6mg produce interference in the determination of 0.6mg Eu, whereas other lanthanides cause no interference. For the determination of Nd, Ho and Er in Longnan (China) rare earth mixtures, the relative standard deviation is less than 1.4 per cent. Ren Ying et al. [25-27] studied the derivative spectra of the rare earth complexes formed with three β-diketones in detail and compared their sensitivities for the determination of Pr, Nd, Sm, Eu, Ho, Er and Tm. They used the third-order derivative spectra

to determine Nd, Er, Ho and Tm directly in a mixture of 15 rare earth elements by use of the RE-TTA system; the relative error was less than 4 per cent [28]. Replacing acetone with Triton X-100 and using multiwavelength-accumulating derivative spectrophotometry to treat these derivative spectra, Li Hinhe et al. [29] obtained the third-order derivative molar absorptivities for complexes formed by Nd, Ho and Er with DBM to be 2288, 4768 and 1408 $l.mol^{-1}.cm^{-1}.nm^{-3}$, respectively. The method is quite accurate and the detection limits are greatly reduced. The third-order and fourth-order derivative spectra of complexes of rare earths with Tiron in 0.05-0.3 M sodium hydroxide solution are reported [30], the proposed method can be used to determine Pr, Nd, Sm, Dy, Ho, Er and Tm directly in rare earth mixtures with satisfactory results. Gao et al. [31] determined Pr, Nd, Sm, Eu, Ho and Er in mixed rare earths by means of fourth-order derivative spectra of complexes of rare earths with PMBP (or TTA) and γ-methylpyridine (or 2,4,6-trimethylpyridine). Bei et al. [32] also studed the absorption spectra of rare earth complexes with TTA and 4-methylquinoline (or 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline). In the concentration rate 0-1000μg/ml of Nd, Ho and Er can be determined. Zhang et al. [33] studied the characteristic absorption spectra of rare earth complexes with PMDCP and DPG, which can be used for determination of Nd, Ho and in rare earth mixtures.

8-Hydroxyquinoline group:

Wang et al. studied systematically the absorption spectra of 4f electron transitions of rare earth complexes with 8-hydroxyquinoline and its derivatives and various reagents/surfactants. The absorption characteristics of those complexes and their application are shown in Table 1.

Inorganic salts

There have been many reports on the absorption spectra of rare earths perchlorate solutions. Ren Ying et al. selected the third-order derivative spectra to determine Pr, Nd, Sm, Eu, Ho, Er, Dy and Tm in mixtures of lanthanides [44], Pr and Nd in Fe-Pr-Nd alloy [45], Pr and Sm in Pr-Sm-Co alloy [46], Gd in mixed rare earths [47], Yb and Dy

Table 1.
Spectroscopic Characteristics of Rare Earth Complexes with 8-Hydroxy-quinoline Group Reagents and Application

Element	Reagent	Acidity (pH)	Wavelength (nm)	Linear Range	Ref
Nd, Er	ClHQ	9.0	581.8 Nd 520..8 Er	0 - 7.0x10^{-4} M 0 - 15x10^{-4} M	34
Nd	HQS - Ninhydrin	6.3	589	0 - 80 μg/ml	35
Pr, Ho	HQS - CPC	5-11	491(+)-485(-)Pr 454(+)-449(-)Ho	0 - 14 μg/ml 0 - 16 μg/ml	36
Nd, Ho	HQS - CPC	6.5	576.8(+)-573(-)Nd 522.5(+)-520.5(-)Er	0 - 25 μg/ml 0 - 30 μg/ml	37

Nd, Pr, Er	HQS - Ninhydrin	7.4	591(+)-588(-)Nd 490(+)-486(-)Pr 523(+)-520(-)Er	0 - 0.48 mg/ml 0 - 0.48 mg/ml 0 - 0.48 mg/ml	38
Nd, Er	Ferron - DEA	alkaline solution	575.4(+)-572.6(-)Nd 521.5(+)-519(-)Er	0 - 13 μg/ml 0 - 13 μg/ml	39
Nd	HQ - Triton X-100	0.4-1.25 M NaOH	585.5(+)-577(-)	0 - 14 μg/ml	40
Nd, Er	HQS - DEA	alkaline solution	575(+)-571.5(-)Nd 521(+)-518(-)Er	0 - 11.8 μg/ml 0 - 12.3 μg/ml	41
Nd, Er	BrHQ-TritonX-100	0.72-1.5 M NaOH	575.5(+)-572(-)Nd 520.5(+)-517(-)Er	0 - 18 μg/ml 0 - 21 μg/ml	42
Nd, Er	HQ-DEA-ethanol	alkaline solution	575(+)-571.5(-)Nd 522(+)-518.5(-)Er	3.0 - 18 μg/ml 4.5 - 21 μg/ml	43

in mixed rare earths [49], and Pr, Ce and Nd in light rare earths and aluminum [48], respectively. Ishii et al. [50] determined Pr in mixed rare earths by the second-order derivative spectra of rare earth perchlorate solutions, where the interference of other lanthanides was eliminated by the isosbestic point found in their derivative spectra. Aleksandrova et al. [51] reported a method for determination of Gd by means of second-order derivative spectra of gadolinium chloride, 10-300-fold concentrations of Sm, Tb, Dy, Ho and Tm did not interfere. In enriched gadolinium nitrate solution, the characteristic absorption peak of Gd at 272 nm is determined by second-order derivative spectra. The precision is 9.038g/l for the determination of 8g/l Gd, the detection limit is 0.071g/l [52]. The characteristic peaks of Nd, Ho and Er are enhanced in 1.5 M K_2CO_3 solution, their molar absorptivities are 36, 24 and 18 $l.mol^{-1}.cm^{-1}$, respectively. The composition of the Nd-CO_3^{2-} complex determined is 1:4 [53].

Other reagents:

Rare earths can form 1:2 and 2:3 complexes with Tiron at pH 5, the characteristic peak of Nd is split with increasing Tiron concentration at pH 12 and the sensitivity is increased. The molar absorptivities for Nd (pH 12), Ho(pH 5.0) and Er(pH 5.0) are 9, 9 and 5 times higher than those of chlorides respectively. The systems follow Beer's Law up to 4mg/ml (Nd, pH 4.5, 578 nm), 2.5mg/ml (Nd, pH 12, 571nm), 4mg (Ho, pH 4.8, 450nm) and 5mg/ml (Er, pH 4.8, 376nm) [54]. Zhou et al. [55] discovered that kojic acid can form stable complexes with rare earths in weakly alkaline solution, the absorption peaks of Nd, Ho and Er in the visible region are enhanced. The third-order derivative molar absorptivities obtained for Nd, Ho and Er are 166, 292 and 190 $l.mol^{-1}.cm^{-1}.nm^{-3}$, respectively. The relative error is less than 1.5 per cent in the determination of Nd in a reference material. Maeck et al. [56] reported that the characteristic peaks of Nd, Sm, Ho and Er are enhanced when rare earths are extracted from an aluminum nitrate salted aqueous phase into an organic medium which contains tetrabutylammonium nitrate, the proposed method is used to determine Nd, Er, Sm and Ho in lanthanide mixtures. Yoshimura et al. [57] discovered that the absorption for Nd at 740.5

nm is increased with the neodymium ion is exchanged to Muromac 50W-X_{12} cation resin. Wang [58] has discovered that the characteristic peak of 4f electron transitions of neodymium with 2-(5-bromo-2-pyridylazo)-5-diethylaminophenol(5-Br-PADAP) and Triton X-100 system is enhanced. The fourth-order derivative molar absorptivities obtained are 5200 $l.mol^{-1}.cm^{-1}.nm^{-4}$. The system follows Beer's Law up to 0.18mμ/ml of Nd. The method provides for the simultaneous determination of Nd and Pr in binary mixtures. Wang et al. [59, 60] also studied the derivative spectra of neodymium complexes with semi-xylenol orange (or xylenol orange) and cetylpyridinium chloride (or cetylpyridinium chloride and Triton X-100). The characteristic absorption of 4f electron transitions of the complex is 350 times that of neodymium chloride. The fourth-order derivative spectra has been used to eliminate the interference of the other lanthanides, and to increase the sensitivity by a further factor of 6. Beer's Law is obeyed for 0-7.0μg/25ml of Nd. The detection limits is 5.8ng/ml. The proposed method has been used for the determination of Nd in mixed rare earths, with satisfactory results.

Fluorimetry

Luminescence spectroscopy is widely used in the study of rare earth ions, in which the binary or ternary complex of terbium, dysprosium, samarium and europium emitted ionic narrow-band lines. The strongest emissions are invariably observed in the $^5D_0 \rightarrow ^7F_1$ and $^7F_2$ transition regions for europium(III), $^4F_{5/2} \rightarrow ^6H_{9/2}$, $^6H_{5/2}$ and $^6H_{7/2}$ transition regions for samarium(III), $^5D_4 \rightarrow ^7F_5$ and $^7F_6$ transition regions for terbium(III) and $^4F_{9/2} \rightarrow ^6H_{15/2}$ and $^6H_{13/2}$ transition regions for dysprosium(III). In Table 2 are listed those characteristics of Sm, Eu, Tb and Dy emission intensity spectra.

Huang et al. studied the fluorimetry of the complex of terbium and dysprosium with salicylic acid and ethylenediaminotetraacetic acid (EDTA)[61], and with dicarboxylic acid such as oxalic acid [62]. The lower limit for the determination of terbium and dysprosium is 6.4 and 30 ppb, respectively. This method was applied to determine micro amounts of terbium and/or dysprosium in rare earth oxides.

Table 2.
Major Characteristics of Sm^{3+}, Eu^{3+}, Tb^{3+} and Dy^{3+} Emission Intensity Spectra for Complexes in Aqueous Solution

Transitions	Spectral Region (nm)	Relative Intensities
Samarium		
$^4F_{5/2} \rightarrow {}^6H_{5/2}$	555 - 570	strong
$\rightarrow {}^6H_{7/2}$	590 - 610	strong
$\rightarrow {}^6H_{9/2}$	640 - 655	strongest
Europium		
$^5D_0 \rightarrow {}^7F_0$	578 - 580	weak
$\rightarrow {}^7F_1$	585 - 600	strong
$\rightarrow {}^7F_2$	610 - 630	strongest
$\rightarrow {}^7F_3$	645 - 660	weak
$\rightarrow {}^7F_4$	680 - 705	weak
Terbium		
$^5D_4 \rightarrow {}^7F_6$	485 - 500	strong
$\rightarrow {}^7F_5$	540 - 555	strongest
$\rightarrow {}^7F_4$	580 - 595	medium
$\rightarrow {}^7F_3$	615 - 625	weak
$\rightarrow {}^7F_2$	645 - 655	very weak
Dysprosium		
$^4F_{9/2} \rightarrow {}^6H_{15/2}$	480 - 495	strong
$\rightarrow {}^6H_{13/2}$	575 - 590	strongest
$\rightarrow {}^6H_{11/2}$	640 - 655	weak

Bel'tyukova et al. have researched the fluorescent reaction of terbium and dysprosium with nalidixic acid by extraction using hexane [63], and terbium, dysprosium and europium with imidazole-4,5-dicarboxylic acid [64]. The lower limit for the determination of terbium, dysprosium and europium is 10 ppb, 200 ppb and 100 ppm, respectively. The method was used for the determination of terbium, dysprosium and europium in rare earth oxides and yttria. They studied the fluorescence characteristics of the complex of terbium and dysprosium with tetraemthylester of 2,6-dioxobicyclo [3,3,1] nonane-1,3,5,7-tetracarboxylic acid [65]. Ci et al. studied the fluorimetric method for the acetylacetone complex of terbium [66, 67], and the dipicolinic acid complex of terbium [68]. The detection limit is 4ng/ml. This method was applied to determine micro amounts of Tb in rare earth oxide and LaOBr. The fluorescence characteristics of the complex of dysprosium with pyrocatechol-3,5-disulphonic acid (Tiron) and EDTA was reported [69]. The fluorescence intensity was increased 22-fold in presence of surfactants. The lower limit for the determination of Dy is 1.0×10^{-9} M, and was used for the determination of Dy in rare earth oxides. Huang et al. studied the fluorescence of terbium in presence of gelatin. The linear range for the determination of Tb is 1-10μg/ml and was used for the determination of Tb in yttrium oxide, lanthanum oxide and gadolinium oxide [70]. Aihara et al. have researched the flow-injection spectrofluorimetric method of the pivaloyltrifluoroacetone (PTA) complex [71] of terbium in presence of trioctylphosphine (TOPO) and nona(oxyethene)dodecyl ether (BL-9EX), and the ethylenediamine bis-(o-hydroxyphenylacetic acid) complex of terbium [72]. The linear range for the determination of terbium was 16-160ng/ml. Lyle et al. have investigated the flow-injection fluorimetric method of the Tiron complex of terbium [73] and dysprosium [74] in presence of EDTA, and this was used for the determination of terbium and dysprosium in rare earth oxides. Ci et al. studied the fluorescence reaction of dysprosium(III) with pyrocatechol-3,5-disulphonic acid (Tiron) and alkaline-earth metal ions, such as Ca^{2+}, Sr^{2+} and Ba^{2+}. In presence of the above alkaline-earth metal ions, the fluorescence intensity of the dysprosium-Tiron complex is 2-3 fold greater [75, 76]. Yang [77], Zhu [78] and Ci [79] found and

investigated systematically a new fluorescence enhancement phenomenon, called as co-luminescence or co-fluorescence. In the presence of lanthanum(III), gadolinium(III), terbium(III), lutetium(III) or yttrium(III), the fluorescence intensity of europium and/or samarium-thenoyl-trifluoroacetone(TTA)-phenanthroline(Phen) (or diphenylphenanthroline + TOPO) system can be increased by 2-3 orders of magnitude. This fluorescence enhancement phenomenon was also reported in europium and/or samarium-benzoylacetone(BA) [81] and europium and/or samarium-dibenzoylmethane(DBM) [80] systems by adding lanthanum, gadolinium, terbium, lutetium or yttrium.

Zhu et al. researched the simultaneous spectrofluorimetric determination of terbium, samarium and europium [82] using the hexafluoroacetylacetone(HFA)-TOPO system in presence of Triton X-100. The lower limit for the determination of terbium, samarium and europium is 5nM, 0.06μM and 5nM, respectively.

Si et al. reported the 2-(diphenylacetyl)indan-1,3-dione (diphacinone,DPN) as a new reagent for the fluorimetric determination of micro amounts of europium in rare earth oxides [83]. The complexes formed by the reaction of samarium ion with DPN and TOPO in the presence of Triton X-100 are fluorescent, and based on this the diphacinone can be determined in serum an durine [84].

Li et al. studied the three-dimensional fluorimetric method for the complex formed by the reaction of terbium, dysprosium, samarium and europium with hexafluoroacetylacetone as the primary ligand, trioctylphine oxide as the synergistic ligand and Triton X-100 as the detergent. The lower limit for the determination of terbium, dysprosium, samarium and europium is 0.5nM, 0.08μM, 5nM and 0.3nM, respectively [85]. The fluorimetric methods of some rare earths are given in Table 3.

Table 3.
Fluorimetric Method of Some Rare Earth Elements to be Determined

Element	Reagent	Acidity (pH)	λex/λem (nm)	Detection Limit	Linear Range	Ref
Tb	Dicarboxylic acid	8.0	266/546	0.03ppm		62
	$H_2SO_4(H_3PO_4)$		222/494	33ng/ml		86
	HFA-TOPO-TX-100	2.6-3.3	320/545	$1.0x10^{-9}$M	$5.0x10^{-9}$-$4.6x10^{-6}$M	87
	Phenylanthranilic acid	7.5-8.5	286/543	20 μg/ml		88
	Gelatin	7.0	291/547	1.0 μg/ml		70
	Dipicolinic acid	5-12	280/542	4 ng/ml	4-240 ng/ml	68
	Acetylacetone (AA)	8.0-8.5	301/540	4 ng/ml	0-0.04 μg/ml	66,67
Dy	Tiron-CTMAB-polyacryl-amide-ethanol	12.5	324/577	1nM	$1.0x10^{-8}$-$4.0x10^{-6}$M	89
	Tiron-EDTA-CTMAB	11-12	326/578	$1.0x10^{-9}$M	$1.0x10^{-8}$-$1.0x10^{-5}$M	69
	Tiron-alkaline earth	11	321/576	0.1ng/ml	0-0.04 μg/ml	75,76
	HFA-TOPO-TX-100					90

Table 3., continued (2)

Element	Reagent	Acidity (pH)	λex/λem (nm)	Detection Limit	Linear Range	Ref
Tb, Dy	Salicylic acid-EDTA	13.2	323/577Dy 323/546Tb	160ppbDy 6.4ppbTb		61
	HFA-TOPO-TX-100	3.0		2pMTb 0.5nMDy		91
	Tiron-iminodiacetic acid	12	328/578Dy 328/545Tb	5.0×10^{-10}M 5.0×10^{-11}M	7.0×10^{-9}-7.0×10^{-6}M 2.0×10^{-10}-6.0×10^{-6}M	92
	Nalidixic acid	0.1M NaOH	328/570Dy 328/545Tb	200ng/ml 10ng/ml		63
Sm	Gd-DMB-NH_3	8.5-10	406/565	8nM	1.0×10^{-8}-5.0×10^{-7}M	93
	Gd-TTA-Phen-TX-100	4.5-8.5	368/565	0.75nM		94,114
	La-TTA-Phen-TX-100	5.8-7.0	370/565	7.5×10^{-10}M	1.0×10^{-8}-8.0×10^{-6}M	95
	Tb-TTA-Phen-TX-100	5.5-7.0	372/565	2.0×10^{-11}M	1.0×10^{-9}-1.5×10^{-7}M	96
	Y-TTA-Phen-TX-100	6.0-9.5	368/565	1.0×10^{-9}M	1.0×10^{-9}-6.0×10^{-8}M	97
	Lu-TTA-Phen-TX-100	6.1-7.3	375/565	4.0×10^{-9}M	1.0×10^{-8}-9.0×10^{-6}M	98
	Tb-TTA-TOPO-TX-100	6.5-8.5	355/562	1.0×10^{-10}M	6.0×10^{-9}-1.0×10^{-6}M	99

Table 3., continued (3)

Element	Reagent	Acidity (pH)	λex/λem (nm)	Detection Limit	Linear Range	Ref
Eu	BTA-TOPO	6.0	335/620	1ppb		100
	TTA-1,10-diaza-15-crown-5	6.5-7.5	365/612	5ppb		101
	TTA-Phen	5.0	374/613	0.02ppb		102
	TTA-diphenylguanidine	6.5	374/611	0.1 μg/ml	0-3.5 μg/10ml	103
	TTA-N'-(2-hydroxy ethyl)-ethylenediamine-acetic acid	8.3	365/613	0.05 μg/25ml	0.05-8.0μg/25ml	104
	2-Propionylindan-1,3 dione	4.0	330/610	0.1ppm		105

Table 3., continued (4)

Element	Reagent	Acidity (pH)	λex/λem (nm)	Detection Limit	Linear Range	Ref
Eu	Diphacinone-TX-100					81
	TTA-CTMAB-TX-100	5.5-7.5	370/612	2.0×10^{-12}M	1.0×10^{-9}-1.0×10^{-7}M	106
	La-TTA-Phen-Tween-20	6.0-6.5	355/612	2.0×10^{-12}M	1.0×10^{-10}-2.0×10^{-7}M	111
	Lu-TTA-Phen-TX-100	6.0-9.0	371/612	5.0nM	5.0×10^{-9}-5.0×10^{-7}M	107
	Tb-TTA-Phen-TX-100	7.0	371/612	0.1pM		108
	Gd-TTA-Phen-TX-100	5.8-7.2	371/612	7.5pM	5.0×10^{-11}-7.0×10^{-7}M	109
	La-TTA-TOPO-TX-100	6.4-8.5	360/617	6.0×10^{-11}M	1.0×10^{-10}-4.5×10^{-7}M	110
	Gd-TTA-TOPO-TX-100	6.0	343/615	5pM	1.0×10^{-10}-1.0×10^{-7}M	112,113
	Gd-TTA-diphphen-TX-100	5.3	346/613	0.6ng/ml	0 - 1.59ng/ml	79
	Y-TTA-Phen-TX-100	5.5-7.5	371/612	1.0×10^{-12}M	1.0×10^{-10}-3.0×10^{-7}M	115
	Gd-TTA-CTMAB-TX-100	7.5-8.5	370/612	2.0×10^{-12}M	6.0×10^{-11}-1.0×10^{-7}M	116

Table 3., continued (5)

Element	Reagent	Acidity (pH)	λex/λem (nm)	Detection Limit	Linear Range	Ref
Eu	La-DBM-diphenyl-guanidine(DPG)	9.0-10	395/612	$5.0x10^{-11}$M		117
	Tb-DBM-DPG	6.5-7.5	390/612	$5.0x10^{-11}$M	$5.0x10^{-10}$-$6.0x10^{-8}$M	118
	Y-DBM-DPG	7.0	390/612	$5.0x10^{-13}$M	$1.0x10^{-10}$-$5.0x10^{-8}$M	119
	Lu-DBM-DEA	9.5-11.5	390/612	$5.0x10^{-12}$M	$5.0x10^{-11}$-$1.0x10^{-7}$M	120
	Tb-TTA-CTMAB	5.5-8.0	372/612	$1.0x10^{-13}$M	$1.0x10^{-12}$-$1.0x10^{-8}$M	120
	Tb-DBM-NH_3	9.0	400/612	$4.0x10^{-11}$M	$1.0x10^{-9}$-$5.0x10^{-7}$M	80
	Y-DBM-NH_3	8.9-9.5	400/612	0.1nM	$1.0x10^{-9}$-$4.0x10^{-8}$M	121
	Gd-DBM-NH_3	9.0-9.8	400/612	$3.0x10^{-11}$M	$1.0x10^{-10}$-$6.0x10^{-8}$M	122
	Tb-BA-Phen-TX-100	8.0	370/612	20pM	$1.0x10^{-9}$-$1.0x10^{-7}$M	81
	Y-DPN-NH_3-TX-100	9.0	330/612	80fM	$6.0x10^{-11}$-$8.0x10^{-7}$M	123
	Gd-DPN-NH_3-TX-100	9.0	330/612	60fM	$1.0x10^{-11}$-$8.0x10^{-9}$M	124
	La-DPN-TX-100	7.0-10	330/612	$3.0x10^{-12}$M	$1.0x10^{-10}$-$8.0x10^{-8}$M	125

Table 3., continued (6)

Element	Reagent	Acidity (pH)	λex/λem (nm)	Detection Limit	Linear Range	Ref
Sm, Eu	TTA-TOPO	5.5	348/644Sm	2ppb		126,127
			348/611Eu	0.02ppb		
	TTA-Phen-polysorbate-20	5.5	345/649Sm	7.0ppb		128
			345/614Eu	0.3ppb		
	TTA-Phen-Tween-80	5.5	345/652Sm	5.6ppb	0-2000ppb	129
			345/616Eu	0.5ppb	0-750ppb	
	Tb-DBM-CTMAB-TX-100	9.0	390/565Sm	1.0×10^{-9}M		131
			390/612Eu	1.0×10^{-12}M		
	Y-TTA-Phen-TX-100	5.2-5.8	370/565/Sm	3.0×10^{-10}M	4.0×10^{-10}-1.0×10^{-7}M	132
			370/612Eu	2.0×10^{-12}M	2.0×10^{-12}-8.0×10^{-9}M	
	Gd-DBM-DEA	9.6	390/650Sm	5.0×10^{-13}M	1.0×10^{-9}-8.0×10^{-8}M	133
			390/612Eu	8.0×10^{-14}M	1.0×10^{-11}-4.0×10^{-9}M	

	Tb-TTA-TOPO-TX-100	7.0	360/565Sm	$2.0x10^{-10}$M	$1.0x10^{-8}$-8.0x10--7M	134
			360/612Eu	$1.0x10^{-10}$M	$1.0x10^{-9}$-$8.0x10^{-8}$M	
	Gd-TTA-Phen-TX-100	5.5-8.2	370/565Sm	$5.0x10^{-10}$M	$2.0x10^{-9}$-$8.0x10^{-8}$M	135
			370/612Eu	$7.5x10^{-12}$M	$5.0x10^{-11}$-$4.0x10^{-9}$M	
Sm	TTA-DPG		345/645Sm	10ppm		136
Eu			345/614Eu	0.5ppm		
Tb			345/545Tb	100ppm		
	PTA-TOPO-BL-9EX	4.5	309/598Sm	150ng/ml		130
			310/611Eu	10ng/ml		
			309/542Tb	10ng/ml		
	HFA-TOPO-TX-100	3.0	330/565Sm	0.06μM	$1.0x10^{-6}$-$1.0x10^{-4}$M	80
			330/612Eu	5nM	$1.0x10^{-8}$-$1.0x10^{-6}$M	
			330/545Tb	5nM	$1.0x10^{-8}$-$1.0x10^{-6}$M	

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