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Review

Energy transfer in solution of lanthanide complexes

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Abstract

The lanthanides with their well-defined energy levels provide an excellent basis to study different Ln(III)-specific energy transfer processes in a variety of chemical environments. The studies concerning intramolecular and intermolecular energy transfer processes with participation of Ln(III) ions and a variety of ligand groups in solution are reviewed. Phenomena of energy transfer from ligands to Ln(III) ions, resulting consequently in a great enhancement of the Ln(III) ion luminescence (ligand sensitized luminescence), as well as from Ln(III) to other species and between Ln(III) ions are presented. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

In the last decade, interest of energy transfer processes in solution of lanthanide (Ln) complexes increases due to their potential applications in luminescent assays for biochemistry, liquid lasers, electroluminescence and telecommunication devices as well as for trace determination of lanthanide ions. The lanthanides with their well-defined energy levels provide an excellent basis to study the different Ln(III)-specific energy transfer processes in a variety of chemical environments. It particularly concerns their application in ultrasensitive bioanalytical assays [1–3] and in optical amplification [4] as well as application to study of coordination compounds in solution [5,6] and as luminescent lanthanide sensors for pH, pO_2 and selected anions [7].

In 1990, an interesting review [8] on quenching of excited states by lanthanide ions and chelates in solution was published in which mechanisms, history and scope of studies on energy transfer processes with the participation of Ln(III) ions have been described. Therefore, in this review, we present studies published, starting from 1989/1990, considering both energy transfer from ligands to Ln(III) ions as well as using Ln(III) as energy donors.

2. Ligands as energy donors

The main effect of participation of the both intramolecular and intermolecular energy transfer in the lanthanide systems is a great enhancement of the lanthanide(III) ion luminescence (ligand sensitized luminescence). Ln(III) ions are chelated with ligands that have broad intense absorption bands. In these systems, intense ion luminescence originates from the intramolecular energy transfer through the excited state of the ligand to the emitting level of the Ln(III) ion. Lehn named this phenomenon the “antenna effect” and proposed that such complexes could be seen as light conversion molecular devices because they are able to transform light absorbed by the ligand into light emitted by the ions via an intramolecular energy transfer [9].

Numerous ligands applied in published papers, we have divided into several groups in order to make easier a description of large number of publications.

2.1. β-Diketones

The effect of the nature of β-diketones (with many various aryl and fluorine alkyl substituents) on the spectrochemical and luminescence properties of lanthanide complexes was examined. It was stated that the charge distribution in the chelate ring, the spatial structure and the hydrophobicity of the coordinated ligand determine the efficiency of energy transfer from the β-diketone to the Ln(III) (Yb, Eu) ion [10].

The investigation of luminescence and energy transfer efficiency in the europium–thenoyltrifluoroacetone (TTA) chelate in ethanol and in aqueous micellar solution was conducted [11]. The method of the fluorimetric determination of europium (with detection limit of 6×10^{-12} mol/l) in its ternary complex with TTA and trioctylphosphine oxide (TOPO) in a micellar solution of Triton X-100 was proposed

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[12]. The luminescence of Eu(III)–TTA-surfactant (Triton X-100 or sodium dodecyl sulphate, SDS) systems was investigated and it was concluded that in the energy transfer process occurring in the micellar structure, two excited states $^5D_1$ and $^5D_0$ play an important role [13]. The soluble complexes $LiEu(TTA)_4$, $NaEu(TTA)_4$ and $KEu(TTA)_4$ luminescent in different solvents and in poly(*N*-vinylcarbazole) thin films, were applied as emitter materials in high-performance light-emitting diodes [14]. The Eu–TTA–CTMAB–Triton X-100 system (CTMAB = cetyltrimethylammonium bromide) was used for determination of Eu(III) with detection limit of 2.2×10^{-12} mol/l [15]. The energy transfer from the excited nitrogen in the triplet state to the triplet state of the TTA ligand and subsequently to Eu(III) was observed during chemiluminescent oxidation of hydrazine (in $[Eu(TTA)_2N_2H_4]$ complex) by hypochlorite [16].

By adding 1,10-phenanthroline (Phen) bounded to the Eu(III)/TTA complex in aqueous micellar solution the luminescence intensity of Eu(III) can be dramatically increased. Both the TTA and Phen molecules absorb the excitation light and the excitation energy from the TTA is transferred partly towards the Phen. Thus, the energy from the TTA triplet level can reach the Eu(III) ions with higher probability in the presence of Phen [17]. Two ternary systems containing Eu(III)–TTA and dinaphthyl-diphenyl-3,6-dioxaoctanediamide (DDD) or trisalicylicamido-triethylamine (TSTA) were used for determination of the Eu(III) ions in rare earth ores and high purity rare earth oxides [18,19]. DDD and TSTA sensitize the luminescence of Eu(III) binary complex with TTA through intramolecular energy transfer from the secondary ligand to the first ligand (TTA). Detection limits were 2.279×10^{-4} μg/ml (original luminescence) and 8.566×10^{-5} μg/ml (first derivative luminescence signal) in the DDD system [18] and 1.520×10^{-8} g/l in the TSTA system [19].

Synergistic enhancement of the luminescence of Eu(III) and Sm(III) by formation of the ternary complexes with 2-naphthoyltrifluoroacetone (NFA) or TTA and TOPO in single-phase oil–water microemulsion droplets composed of water–SDS–butan-1-ol-heptane was observed as described in [20]. In dissociation-enhanced lanthanide fluoroimmunoassay (DELFIA), NFA is used (with detergent and TOPO) as an enhancement solution giving a chelate with Eu(III) which emits intense luminescence [21]. The method of simultaneous spectrofluorimetric determination of Sm(III), Eu(III) and Tb(III) with hexafluoroacetone, TOPO and Triton X-100 was presented, with detection limits 6.0×10^{-7} mol/l for Sm(III), 8.0×10^{-9} mol/l for Tb(III) and 5.0×10^{-9} mol/l for Eu(III) [22].

The energy transfer processes from different organic compounds to lanthanide (mainly Tb(III) and Gd(III)) acetylacetonates, to the ligand-localized triplet states as well as to the ff* states of lanthanide ions were investigated [23]. The fundamental role of the ground state Tb(III)/acetylacetone (1:1) complex in sensitized emission of Tb(III) in ethanol solution was discussed in [24]. The luminescence enhancement by electron-withdrawing groups in β-diketones, after excitation of these groups, in Eu(III)–β-diketonato–TOPO ternary complexes was investigated. The Eu(III) luminescence quantum yield changes with kind of substituent groups as follows: $C_4F_9 \geq C_3F_7 > C_2F_5 > CF_3$. These complexes can be used in the Eu(III) determination with the detection limit of the complex $[EuL_3(TOPO)_{1.5}]$ (L = heptafluoro-phenanthryl-4,6-hexanedione) equal to 4.42×10^{-12} mol/l [25].

The luminescence intensity enhancement of Ln(III) (Sm, Eu, Nd, Yb) β-diketone (acetylacetone, trifluoroacetone, benzoylacetone) copolymers with styrene or methyl methacrylate was also observed [26,27]. It was found that its intensity depends not only on the character of the substituent in the β-diketone molecule, but also on the distance between β-diketone fragments in the copolymer [26]. Europium (in the concentration range of 10^{-3}%) was determined in Ce, Pr, Nd and Ho oxides, which are strong luminescence quenchers, using copolymers containing trifluoroacetone and benzoylacetone [27].

The formation of mixed ligand complex after introduction of neutral ligand, i.e. 1,10-phenanthroline into the Yb(III)–β-diketonate molecule (fluorinated thienyl and phenyl derivatives of acetylacetone) improves screening of the central ion providing actually complete transfer of energy, absorbed by the ligand, to the ytterbium ion [28].

The coordination of Michler's ketone (bis(*N*,*N*-dimethylamino)benzophenone) with lanthanide β-diketonate, i.e. europium tris(heptafluoro-2,2-dimethyloctane-3,5-dione) $(Eu(fod)_3)$ in benzene enables efficient sensitization of Eu(III) luminescence after excitation of ligand at wavelengths extending into the visible region (∼450 nm). It is interesting property, e.g. for application as a luminescent label in biological materials, since visible light is less harmful to biological tissue than ultraviolet one [29].

An unexpected effect of enhancement of the $Eu(fod)_3$ luminescence (upon excitation into the absorption band of the ligand) under the action of H_2O (D_2O) molecules in toluene solutions was observed. It is due to the formation of outer-sphere associates $[Eu(fod)_3 \cdot nH_2O]$ by hydrogen bonding of fluorine atom of the ligand with OH-group of water. Data obtained demonstrate the possibilities of the non-radiative energy transfer technique for studying inner- and outer-sphere complex formation in Ln(III) ions in solutions [30]. It was shown that also the luminescence of europium, samarium and terbium β-diketonates (L = benzoyltrifluoroacetone, thenoyltrifluoroacetone, acetylacetone, dipivaloylmethane and heptafluorodimethyloctanedione) in toluene solutions was enhanced by water at $[LnL_3 \cdot nH_2O] > 10^{-4}$ mol/l, while the luminescence of dysprosium, neodymium and ytterbium diketonates was quenched by water. In toluene, the Ln(III) β-diketonates show significant concentration quenching due to the formation of dimers at high concentrations of chelate. The dimers have lower luminescence quantum yields compared to monomers due to energy losses both in the ligand and in the Ln(III) ion. The presence of water in toluene causes dissociation of poorly luminescent dimers to give monomers, which results in unexpected significant enhancement of the luminescence intensity of Eu(III), Sm(III) and Tb(III) β-diketonates. Probably, addition of water enhances ligand to Ln(III) energy transfer efficiency. The luminescence of Dy(III), Nd(III) and Yb(III) β-diketonates is quenched by water due to an efficient deactivation of excited states of these ions by the OH-groups. In the case of Eu(III) chelates, probably water enhances the luminescence quantum yield not only by shifting the equilibrium monomer–dimer, but also by increasing the energy of ligand-to-metal charge-transfer (LMCT) state [31,32].

The addition of yttrium or certain lanthanide ions (lanthanum, gadolinium and lutetium) can enhance the luminescence intensities of the europium (samarium, dysprosium, terbium) β-diketonate when the chelates are present in the form of a suspension in an aqueous solution or in the micellar environment. The enhancing ion must not contain excited 4f levels situated below the excited triplet level of the β-diketone used. This is referred to as a columinescence and is based on an intermolecular energy transfer from the chelates of the enhancing ion (e.g. Y(III)) to the chelates of the emitting ion (such as Eu(III)). As the concentration of the donor is generally much greater than that of the acceptor complex, each acceptor is surrounded by many donor chelates, and the emission of the acceptor is greatly enhanced [33]. The use of the columinescence effect increases lanthanide chelate detectability and also allows Sm(III) and Dy(III) emission measurements in fluoroimmunoassays.

The Sm(III)–TTA–Phen–Gd(III)–Triton X-100 system can be applied to the determination of trace amounts of samarium in lanthanide oxides, with detection limit of 8×10^{-12} mol/l [34]. This same system was used to the simultaneous determination of samarium and europium in mixtures of lanthanides (detection limits: 5×10^{-10} mol/l for samarium; and 7.5×10^{-12} mol/l for europium) [35]. For the determination of trace amounts of europium (10^{-12} to 10^{-13} mol/l) in rare earth samples the systems Eu(III)–TTA–CTMAB–Triton X-100 (enhancement by Gd(III)) [36] and Eu(III)–TTA–CTMAB–Triton X-100 (enhancement by adding of Tb(III)) [37] were used. The addition of Tb(III) also enhances the emission of Eu(III) (detection limit of 10^{-13} mol/l) in the Eu(III)–TTA–dibenzo-18 crown 6–Triton X-100 system what enables the determination of trace amounts of europium in lanthanum, praseodymium and dysprosium oxides [38]. Similarly, for the determination of trace amounts of europium in yttrium and gadolinium oxides, the system TTA–TOPO–Triton X-100 with the excess of Tb(III) was proposed [39].

In the presence of an excess of Y(III), the luminescence intensities of Eu(III) and Sm(III) chelated with benzoyltrifluoroacetone (BTA) or TTA in an aqueous solution containing 1,10-phenanthroline and Triton X-100, were increased several-hundred times. This phenomenon was used in the time-resolved fluorimetric determination of Eu(III) and Sm(III) as well as in the double-label time-resolved fluorimetric immunoassay of luteinizing hormone and follicle stimulating hormone. The detection limits of Eu(III) in the BTA- and TTA-based solutions were 4 and 15 fmol/l, respectively. The detection limits of Sm(III) were 0.11 and 0.12 pmol/l, respectively [40]. The luminescence enhancement of Eu(III)–TTA–Phen in micelle solution by adding other Ln(III) (La, Gd, Lu, Y) ions was also investigated [41]. The luminescence enhancement of Eu(III) by Tb(III) in TTA–Phen and BTA–Phen micelle solution was observed as the result of two mechanism: (1) the “wrapping effect” of terbium complexes for europium complexes; and (2) the intermolecular energy transfer from Tb(III) complexes to Eu(III) ions [42]. A method based on the synergistic luminescence of Eu(III) (in the presence of Tb(III) ions) in TTA–Phen–Triton X-100 system was used for the determination of human immunoglobulin G [43]. In the Eu(III)–benzoylacetone–Phen system, after addition of Tb(III) ions the detection limit for europium was 2.0×10^{-11} mol/l [44].

A sensitive columinescence enhancement system was developed for the fluorimetric determination of europium, terbium, samarium and dysprosium ions. The luminescence intensities of the ternary chelates with pivaloyltrifluoroacetone (PTA) and 1,10-phenanthroline in a solution containing Triton X-100 and ethanol were greatly enhanced by the addition of an excess of Y(III), Lu(III), Gd(III) or La(III). Yttrium enhanced the luminescence intensities of Eu(III), Sm(III) and Dy(III) chelates about 100 times and Tb(III) chelate about 1000 times. The detection limits of Eu(III), Tb(III), Sm(III) and Dy(III) were 0.035, 0.34, 7.9 and 46 pmol/l, respectively [45]. Compared to the earlier described solution, the replacement of the synergistic agent 1,10-phenanthroline by 2,2′-bipyridine, and omission of the organic solvent, increased the emission intensity obtained and further improved the detection sensitivities for all of the tested ions (the detection limits for Eu(III), Tb(III), Sm(III) and Dy(III) were 0.019, 0.27, 3.8 and 20 pmol/l, respectively). This system was also tested in dual-label time-resolved immunofluorometric assay of luteinizing and follicle stimulating hormones based on the use of Eu(III) and Tb(III) as the label ions [46].

In the Eu(III)–dibenzoylmethane (DBM)–NH_3 system in the presence of Tb(III), the detection limit of Eu(III) is 4.0×10^{-11} mol/l, which is about two orders of magnitude lower than that of the system in the absence of Tb(III) [47]. A luminescence enhancement produced by adding Gd(III), Y(III), Tb(III), La(III) or Lu(III) to europium (or samarium)–DBM–diethylamine (DEA) system was observed. The gadolinium ions enhanced the luminescence intensity by two to three orders of magnitude compared with the system without Gd(III) and the detection limits were 5×10^{-13} mol/l for Sm(III) and 8×10^{-14} mol/l for Eu(III) [48]. The enhancement effect (by a factor of about 100) on the luminescence intensity of the Eu(III)–DBM–DEA system by lutetium(III) was also observed in a colloidal suspension [37]. The Eu(III)–DBM–cetylpyridinium chloride (CPC) system luminescence intensity can be greatly increased by Gd(III) in the presence of triethanolamine. It was applied to the determination of Eu(III) in rare earth oxides with the detection limit 2.6×10^{-11} mol/l in an aqueous solution [49]. The luminescence of the Sm(III)–DBM–diphenylguanidine system, in the presence of Gd(III) in ethanol–water solution, was applied to the determination of samarium (in the range of 1.0×10^{-9} to 2.0×10^{-6} mol/l) in the different rare earth samples [50].

2.2. Other ligands with carbonyl groups

Efficient energy transfer from steroids containing α-, β-unsaturated carbonyl group (e.g. testosterone, progesterone, cortisone) to Tb(III) ions in micellar media (SDS) permits its determination in biological fluids, in combination with high-performance liquid chromatography [51–53].

Intramolecular energy transfer from tetracycline to Eu(III) ions was used for the sensitive detection of tetracycline in aqueous solution with Triton X-100 [54]. The enhancing effect of TTA on the luminescence intensity of the Eu(III)–tetracycline–Triton X-100 system was observed and used for the determination of tetracycline in serum [55]. A range of micellar system enhancing the Eu(III) luminescence sensitized by a wide range of tetracyclines was investigated [56]. The determination of tetracyclines in aqueous solutions, based on europium luminescence, was improved using EDTA as co-ligand and cetyltrimethylammonium chloride as surfactant [57]. Luminescence enhancement of the Eu(III) in complex with tetracycline (and its derivatives) by DNA was studied. Energy transfer was observed from tetracyclines to Eu(III) both in the presence and in the absence of DNA, which increases the luminescence lifetime and improves the luminescence quantum yield due to decreasing the non-radiative deactivation. Such system was applied to the determination of DNA [58] also in the presence of RNA [59].

Ligand containing three carbonyl groups, 3-acetyl-4-oxy-6-methyl-2-pyrone, transferring the excitation energy to Tb(III) ions, was proposed as a sensitive reagent for the luminescent determination of Tb(III) in lanthanide oxides (with detection limit of 1×10^{-5}% in La_2O_3) [60].

Intramolecular energy transfer in Eu(III) and Tb(III) complexes with Ruhemann's Purple (reaction product of ninhydrin and ninhydrin analogues with amino acids) was studied. It was stated that in this case, the mechanism of intramolecular energy transfer is different than in other Eu(III) and Tb(III) chelates studied in literature. It proceeds, without the participation of the ligand triplet state, by higher excited singlet state of the ligand to higher states of lanthanide ions, and then cascading from these states takes place down to the emitting Ln(III) levels [61].

Sensitized by ligand emission of Eu(III) in the Eu(III)–diphacinone (diphenylacetylindan-1,3-dione)–ammonia system was applied, in the presence of Gd(III) ions, for the determination of europium in rare earth oxides [62] as well as for the determination of rodenticide diphacinone in serum (in the presence of Triton X-100) [63].

Chemiluminescent reaction of adamantylideneadamantane-1,2-dioxetane (AAD) decomposition leds to the formation of singlet and triplet excited adamantanone (Ad=O*). Chemiexcitation of Ln(III) ions in this system might occur by intermolecular and intramolecular energy transfer from Ad=O* [64–71]. In the presence of coordinatively saturated Ln(III) chelates, the excitation of lanthanides takes place through the intermolecular triplet–triplet (also singlet–singlet in the case of TTA and BTA) energy transfer from the excited adamantanone to the ligand (in complex with Ln(III)) and then an intramolecular energy transfer to Ln(III). The coordinatively unsaturated Ln(III) chelates, such as $Ln(fod)_3$ and $Ln(dpm)_3$ (dpm = dipivaloylmethane) form with AAD the ternary complex and the decomposition of AAD (and excitation of Ln(III)) take place in the inner coordination sphere of lanthanide ion through the intramolecular energy transfer from the Ad=O* to the Ln(III) ion. As a result of thermal decomposition of AAD in the complex $[Eu(fod)_3AAD]$, the chemiluminescence (CL) spectra revealed the signal ascribed to the $^5D_1$ level of Eu(III) ion, beside the bands attributed to $^5D_0$–^7F_j transition. The emission from the $^5D_1$ level is attributed to the process of direct (without ligand triplet) transfer of the energy from the excited Ad=O* to the luminescent Eu(III) levels in the geometrically distorted $[Eu(fod)_3Ad{=}O]$ complex [64,65]. Also the spectra of CL accompanying the AAD decomposition in solution of Eu(III) and Tb(III) perchlorates contain the emission bands of these ions due to energy transfer from the excited ketone Ad=O*. This process is more efficient for Tb(III) than for Eu(III) what is related to the difference in the energy gap between the triplet level of Ad=O* and the excited levels of the lanthanide [66]. Similarly, the energy transfer processes were observed as a result of decomposition of AAD in the presence of Eu(III) and Tb(III) tris(benzoyltrifluoroacetonates) [67]. CL of Pr(III) in visible and infrared spectral regions was observed upon decomposition of AAD catalyzed by β-diketonates PrL_3 (L = fod or dpm). The excitation of Pr(III) occurs through intracomplex energy transfer from the excited Ad=O* formed upon decomposition of AAD in the complex $[PrL_3AAD]$ [68–70]. Intermolecular singlet–singlet energy transfer from excited Ad=O*, formed at decomposition of AAD, to the excited level of β-diketonate ligand in Nd(III) and Yb(III) complexes with TTA was studied and infrared CL of Nd(III) and Yb(III) observed. Chemiexcitation of Nd(III) and Yb(III) chelates occurs both at intermolecular and intracomplex mechanisms. With Nd(III) chelates, the efficiency of intracomplex excitation is higher than intermolecular (similarly, as in the case of Pr(III) chelates), while with Yb(III) intermolecular excitation is more efficient than intracomplex one [70,71].

2.3. *Aromatic carboxylic acids*

The aromatic carboxylic acids are also known as ligands transferring energy to the lanthanide ions. Sodium benzoate–TOPO–Triton X-100 system has been shown to enhance the luminescence of terbium, europium and dysprosium in aqueous medium, what is sufficient to lower the detection limits of these lanthanides to the 10^{-9} mol/l level for Tb(III), and 10^{-8} mol/l for Dy(III) and Eu(III) [72]. Luminescence enhancement of Tb(III) (also Dy(III) and Eu(III)) by about three orders of magnitude, due to ligand sensitized luminescence was observed using the following acids as ligands: phthalic (benzene-1,2-dicarboxylic), isophthalic (benzene-1,3-dicarboxylic), terephthalic (benzene-1,4-dicarboxylic), hemimellitic (benzene-1,2,3-tricarboxylic), trimesic (benzene-1,3,5-tricarboxylic), pyromellitic (benzene-1,2,4,5-tetracarboxylic), mellitic (benzene-1,2,3,4,5,6-hexacarboxylic) and *o*-toluic (methylbenzenecarboxylic). This opens up the possibility of the lanthanide determination at trace levels [73].

It was observed that the luminescence intensity of the Tb(III)–trimesic acid–β-cyclo-dextrin system can be obviously increased by $ZrO_3{}^{2-}$ (and some other oxy-ions such as $BiO_3{}^{3-}$ and $SbO_3{}^{3-}$). The authors proposed the mechanism of this enhancement induced by charge transition. The Tb(III) chelate is tightly surrounded by $ZrO_3{}^{2-}$ anion shell which overlapping with 4f orbit of Tb(III) and transferring the charges into it. This transferring action increases the electron density of 4f orbit of Tb(III), so that the efficiency of the energy transfer was increased and greater luminescence was emitted. Such system can be applicated to the determination of Tb(III) (0.508 ng/ml) [74].

It was stated that ligand sensitized luminescence of Tb(III) complexes with methyl-, methoxy-, nitro- and amino-benzoic acids is synergistically enhanced by the addition of TOPO in Triton X-100. The synergism displayed by TOPO was observed only when Triton X-100 micellar medium was used, while the TOPO dissolved in the cationic surfactant dodecyl trimethyl ammonium bromide or anionic one sodium dodecyl sulphate, did not result in synergistic luminescence enhancement of Tb(III). These observations are discussed on the basis of the structure of the surfactant. The observed synergism could be due to the energy transfer from Triton X-100 to Tb(III) complexes. The complexation between TOPO and Tb(III) can result in shortening the distance between the Tb(III) and Triton X-100 compared to that in the absence of TOPO inside the micelle. The reduction of distance between Tb(III) and Triton X-100 can increase the energy transfer efficiency from the phenyl ring of Triton X-100 to Tb(III) resulting in luminescence enhancement of Tb(III) [75].

The luminescent properties of Tb(III) chelated with 1,2- and 1,4-hydroxybenzoic and *o*- and *p*-aminobenzoic acids were also investigated. It was shown that for these benzoic acid derivatives with equivalent triplet state energy levels, sensitization of terbium luminescence is mainly determined by the ability of the acid to bind the lanthanide ion. The luminescence is appreciably enhanced when the –OH and $–NH_2$ groups are in the *ortho* position with respect to the carboxylic group [76]. The determination of anthranilic acid derivative drugs (furosemide, mefenamic and tolfenamic acids) based on the energy transfer from anthranilates to Tb(III) in alkaline methanolic solution was described in [77]. The sensitized Tb(III) luminescence was used for the determination of 1,4-aminobenzoic acid in pharmaceutical preparations [78] as well as for the simultaneous determination of a preservative and a sweetener, namely benzoic acid and saccharin (1,2-sulphobenzoic acid imide), respectively, in food samples [79].

The sensitized emission of terbium in the system Tb(III)–propyl gallate–SDS was used for the determination of antioxidant propyl gallate in edible and cosmetic oils [80]. For the determination of lasalocid, a carboxylic polyether antibiotic used in veterinary practice, the sensitized Tb(III) luminescence was applied [81].

The sensitization of Tb(III) luminescence by the salicylic acid and its derivatives, in ternary chelates with ethylenediaminetetraacetic acid (EDTA), was applied to the determination of salicylic, 4-aminosalicylic and 5-fluorosalicylic acids [82] and diflunisal (2,4-difluoro-4-hydroxy-3-biphenylcarboxylic acid) [83]. Such systems were also used for the simultaneous determination of diflunisal and salicylic acid in serum and urine [84,85]. The phosphate derivatives of salicylic acid were used in the enzyme-amplified lanthanide luminescence method for the determination of α-fetoprotein in serum [86,87]. In [87] 14 ligands (salicylic acid, quinolones and hydroxybenzenesulphonic acid derivatives) were also examined as efficient energy donating chelators for Tb(III) in ternary system with EDTA. The ternary complex of Tb(III) with diethylenetriaminepentaacetic acid (DTPA) and 4-aminosalicylic acid was used as a label for the determination of human serum albumin [88]. The luminescence of Tb(III)–fluoro-salicylate system combined with enzymatic amplification was applied in DNA hybridization assays [89].

It was demonstrated that di-4,4′-hydroxybenzoic acid, generated in the oxidation reaction of 4-hydroxybenzoic acid by H_2O_2 in the presence of hemin, forms a luminescent complex with Tb(III)–EDTA. This fact was used for the determination of hemin [90]. Ternary complex Tb(III)–EDTA–biphenol chromofore was used for the determination of horseradish peroxidase, which catalyses the dimerization reaction of 4-hydroxyphenylpropionic acid to form a luminescent biphenol [91] and applied for the enzyme-amplified lanthanide luminescence bioassays [92].

The triplet sensitized energy transfer from 2-naphthaleneacetic acid to Tb(III)- and Eu(III)-bond reverse micelles (bis(2-ethylhexyl)sulphosuccinate) was investigated. The bond Ln(III) ions provide a remarkable enhancement of the sensitized luminescence compared to free ions. These results indicate that donor molecules are located at the interface close to the Ln(III) ions which are located on the water side of the water/organic interface. The efficiency of energy transfer is higher for Eu(III)-surfactant compared to Tb(III)-surfactant, while higher sensitization is obtained from the latter [93]. It was found that the energy transfer from an organic analyte donor containing a polar substituent group to a Tb(III) counterion in these same reverse micelles occurs predominantly through a diffusion-controlled mechanism, although the presence of other mechanisms is not completely ruled out. Such system was applied to the determination of theophylline, naphthaleneacetic acid, acetylsalicylic acid and caffeine [94].

The columinescence was observed in complexes of europium (also terbium and dysprosium) with certain aromatic acids, i.e. trimesic, pyromellitic and mellitic. The luminescence of Eu(III) was enhanced by over two orders of magnitude by the addition of La(III) used in excess [95,96]. The mechanism of columinescence in these systems is probably different from that observed with β-diketones, where the intermolecular energy transfer occurs due to close proximity between donor and acceptor complexes [33]. There is a correlation between the columinescence process and the structure of ligand. The addition of TOPO to the Eu(III)–aromatic acid–La(III) system was detrimental to the process of columinescence. These both factors indicate the polynuclear complex formation and intramolecular energy transfer as a possible mechanism for luminescence enhancement of Eu(III) (Tb(III), Dy(III)) in the presence of La(III) [96]. The emission intensities of the Tb(III) were increased up to three to five times when the Ln(III) ions were added into the Tb(III)–benzoic acid ethanol solution in concentration almost equal to that of Tb(III) and it was found that the enhanced Tb(III) luminescence is attributed to energy transfer from the Ln(III) complexes to the Tb(III) complex [41].

The Tb(III) and Eu(III) complexes with lipophilic phenylphosphonic acids show efficient energy transfer luminescence and the energy transfer from Ar-P linkage is about 12–90 times more efficient than that from ArO-P [97]. Luminescence and columinescence enhancement of Tb(III) and Eu(III) with phenylphosphonic (PPOA) and phenylphosphinic (PPIA) acids as ligands were also investigated. Enhanced terbium and europium emission observed in complexes with PPOA and PPIA was due to the ligand sensitized luminescence. TOPO participates in bonding with Tb(III) ion in Tb–PPOA and Tb–PPIA complexes. Addition of TOPO has resulted in further luminescence enhancement of Tb(III) in PPIA complex, no such synergism was seen with PPOA complex. In the Eu(III)–PPOA system the luminescence enhancement was observed after addition of La(III) ions (columinescence), but the presence of TOPO has completely negated this enhancement. In PPOA, both OH-groups are involved in bonding with different lanthanide ions resulting in a polynuclear complex involving La(III), Eu(III) and PPOA. Such a polynuclear complex formation is not possible with PPIA as it contains a lone OH-group. The fact that addition of TOPO has decreased the columinescence enhancement support the possibility of polynuclear complex formation in the PPOA case, since TOPO forming an insulating sheath around the individual Ln(III)-ligand complex inhibits the formation of polynuclear complexes. So, the energy transfer mechanism in columinescence of PPOA complexes is probably different from the intermolecular transfer mode and offers a method for trace level detection of Tb(III) and Eu(III) [98].

The 1,2-bis(2-aminophenoxy)ethanetetraacetate and 1,2-bis(2-amino-5-fluorophenoxy)-ethanetetraacetate can transfer the energy to Tb(III) and Dy(III) ions in aqueous solution, without any addition of surfactant or organic solvents [99]. The energy transfer luminescence of Tb(III), Eu(III) and Dy(III) in complexes with bis(2-hydroxybenzyl)ethylenediaminediacetic acid was observed. The detection limit of Tb(III) in such aqueous solution (without any detergent) was 2.1×10^{-11} mol/l [100]. The chelate of Eu(III) with 2,6-bis[*N*,*N*-bis(carboxymethyl)aminomethyl]-4-benzoylphenol forming a dimeric structure, where two Eu(III) cations are encapsulated into the chelating cage is capable of initiating the sensitized Eu(III) emission [101] and can be used as luminescent label in capillary electrophoresis separation of variety of samples of biological interest [102].

In the CL-generating system Eu(II)/Eu(III)–hydrogen peroxide, where europium is complexed with cyclic and acyclic aminopolycarboxylic acids, the energy transfer from the dimols of singlet oxygen (product of simultaneous reactions of H_2O_2 decomposition and radicals recombination) through aminopolycarboxylate ligand to Eu(III) ions and the enhancement of Eu(III) emission were observed [103,104].

2.4. Terphenyl-based compounds

The lanthanides europium, samarium, terbium, dysprosium, praseodymium, neodymium and ytterbium showed sensitized emission via the *m*-terphenyl part of the organic ligand. The luminescence obtained in this way may be useful for diagnostic multilabel applications and for use in optical amplifiers [105–107]. The luminescence of Eu(III), Tb(III), Dy(III), Sm(III), Nd(III), Yb(III) and Er(III) in complexes with a series of terphenyl-based ligands bearing three coordinating oxyacetate and two amido or two sulfonamido groups, was studied. The near-infrared emitting complexes (neodymium, erbium and ytterbium) exhibit sensitized emission with lifetime in the microsecond time-scale and are very soluble in organic solvent, which is essential for the production of polymer-based optical amplifiers [108].

Terphenyl-based Ln(III) (Eu, Tb, Nd, Yb, Er) complexes with a triphenylene antenna chromophore exhibit sensitized visible and near-infrared emission upon photoexcitation of the triphenylene antenna. It was concluded that intramolecular energy transfer rate is higher in the Eu(III) and Tb(III) complexes than in the near-infrared emitting Nd(III), Er(III) and Yb(III) complexes [109,110].

It was explained that transition metal complexes $[Ru(bpy)_3]^{2+}$ and ferrocene are able to sensitize Nd(III) and Yb(III) luminescence in the systems based on terphenyl-based lanthanide complexes that are covalently linked to the transition metal complexes [111].

Intramolecular energy transfer from the organic dyes to Nd(III) was established in the four terphenyl-based dye-functionalized Nd(III) complexes. The sensitization process is the most efficient in the dansyl complex, followed by the lissamine and Texas Red complexes, and least efficient in the coumarine complex [112].

2.5. Pyridine derivatives

The luminescent properties of the Eu(III) and Tb(III) chelates formed with dipicolinic acid (DPA, pyridine-2,6-dicarboxylic acid) in aqueous solution were studied and it was observed that the energy transfer process is less efficient with terbium than with europium. The comparative study of the luminescent properties of Eu(III) and Tb(III) coordinated with DPA and TTA gives the conclusion that the lanthanide ion luminescence sensitized by the coordination with organic ligands depends on several factors including the number of atoms available from the ligand for coordination and the composition of the chelate [113].

The energy transfer from DPA ligand to Tb(III) was used for the bacterial endospore detection and determination (CaDPA is present in the endospore casing and is release into solution with $TbCl_3$) [114–116]. The 4-substituted derivatives of DPA are also the effective energy donors for Tb(III) and Eu(III) ions. For the labeling of protein molecules in luminescence immunoassays the 4-iodoacetamidodipicolinic acid and Tb(III) were proposed [117]. The spectral properties of Tb(III) complexes with a series of 4-substituted analogues of DPA were compared. The Tb(III) emission intensities were in the following order of substituents: $NH_2 > NHAc > OH > H > Cl \approx Br$. The values of relative quantum yield for energy transfer were ordered as follows: $NH_2 > OH > NHAc > Cl > H \approx Br$. Energy transfer efficiency was not enhanced when Br was substituted for Cl at the 4-position, consistent with energy transfer from excited singlet rather than triplet states [118].

It was demonstrated that 1-hydroxy-2(1*H*)-pyridinone can serve as a useful antenna for energy transfer luminescence of Tb(III) bound to this heterocycle [119]. Eu(III) chelate with 4-(phenylethynyl)-2,6-bis[*N*,*N*-bis(carboxymethyl)aminomethyl]pyridine can be used as luminescent label in capillary electrophoresis separation of biological samples (e.g. amino acids, peptides, proteins) [102].

The binucleating complexing agents, bis(4-pyridine-2,6-dicarboxylic acids), were synthesized and luminescence enhancement of their Eu(III) complexes by Y(III) ions was observed in aqueous solution, without other additives and micelle formation. These ligands, capable of coordinating simultaneously two cations, facilitate the formation and growth of a polymeric structure, where Eu(III) and Y(III) ions are linked together through ligands. One Eu(III) ion is able to be excited either by the energy absorbed by three ligands which are coordinated directly to it, or by several other ligands, which transfer their energy, along the polymer chain, to this Eu(III) ion through ligands coordinated directly to it. The Y(III) ion is used (as a non-luminescent ion) to increase both intersystem crossing to ligand triplet level and consequent triplet–triplet energy transfer between ligands [120].

The efficient energy transfer from ligand to metal was observed for the Eu(III) complex with 2,2′-bipyridine-6,6′-dicarboxylic acid [121]. The emission of europium and terbium complexes with 2,2′-bipyridine in poly(ethylene glycol)-200 aqueous solution was investigated. It was observed that the excitation at the bipyridine ligand absorption wavelength resulted in ligand to metal energy transfer and strong luminescence of the lanthanide ions. These complexes, stabilized by association with the polymer chain, may be used to obtain the transparent organic/inorganic matrixes by the sol–gel method with the tetramethoxysilane [122].

The luminescent properties of many different chelates, which can be applied as luminescent labels in fluoroimmunoassays, were presented. These chelates are composed of bipyridine, terpyridine and (arylethynyl)pyridine derivatives as the energy absorbing and donating groups, Eu(III) and Tb(III) as the emitting ions and methylene-nitrilotriacetic acids as the stable chelate-forming moieties [123–127]. The lanthanide complexes of polyacid ligands derived from 2,6-bis(pyrazol-1-yl)pyridine, pyrazine and 6,6′-bis(pyrazol-1-yl)-2,2′-bipyridine that also may be used as lanthanide (Eu, Tb, Sm, Dy) markers in bioaffinity assays were described [128]. The luminescent Tb(III) complex with [2,6-bis(3′-aminomethyl-1′-pyrazolyl)-4-phenylpyridine] tetrakis(acetic acid) was used in fluoroimmunoassay of α-fetoprotein and carcinoembryonic antigen in human serum [129]. The Eu(III) chelate with the terpyridyl group as sensitizer, which can incorporate an isothiocyanate group for biological labeling was described [130]. The Eu(III) complex with terpyridine-bis(methyleneamine)tetraacetic acid was used for the determination of nucleic acids and antibodies [131].

2.6. Quinoline and phenanthroline derivatives

Efficient intramolecular energy transfer from the lowest triplet state of 8-quinolinato moiety to the Nd(III) ion in tris(8-quinolinato)neodymium(III) was investigated. Introduction of halogen atoms in the ligand led to enhancement of the near-infrared Nd(III) luminescence, under visible light excitation. This phenomenon should be explained by increasing of the intersystem crossing from the excited singlet state to the excited lowest triplet state of the ligand and as well as by improvement of the energy overlapping between the lowest triplet state of ligands and the excited levels of Nd(III) [132,133]. The emission from Sm(III) and Eu(III) is greatly sensitized due to intramolecular energy transfer in aqueous solution of complexes with the 2[(2-amino-5-methylphenoxy)methyl]-6-methoxy-8-aminoquinolinetetraacetate [99]. The intrasystem energy transfer in complexes of Eu(III) with 3,3′-biisoquinoline-2,2′-dioxide and with cryptate incorporating this species was described in [134].

Fluoroquinolones (synthetic antibacterial agents) sensitize the lanthanide luminescence as a result of intramolecular energy transfer between the quinolone and lanthanide ions. The emission of Tb(III) complexed with ciprofloxacin was used for the liquid chromatographic determination of this antibiotic in biological fluids [135]. Using Tb(III) luminescence measurements, the determination of ciprofloxacin and enrofloxacin in edible and animal tissues [136] as well as levofloxacin in pharmaceutical samples, human urine and serum [137] were described. It was observed that the sensitized luminescence of Tb(III) in the presence of sparfloxacin is further enhanced, when this system is exposed to 365 nm light. Irradiation of the Tb(III)–sparfloxacin complex causes photochemical reactions taking place in the ligand part. The product forms a new complex with Tb(III), which is more favorable to the intramolecular energy transfer. On this basis, a sensitive and selective method of sparfloxacin determination was established [138]. The photochemical luminescence enhancement was also observed in the case of Tb(III)–lomefloxacin complex [139]. The luminescent properties of Tb(III) and Eu(III) complexes with some quinolonecarboxylic acid derivatives (norfloxacin, ofloxacin, ciprofloxacin, lomefloxacin and perfloxacin) as labels for immunofluorimetric assay were described in [140].

The lanthanide complexes based on two linear (DTPA and TTHA = triethylenetetra-aminehexaacetic acid) and two macrocycle (1,4,7,10-tetraazacyclododecane-tetraacetic and 1,4,8,11-tetraazacyclotetradecanetetraacetic acids) ligands, covalently joined to 7-amino-4-methyl-2(1*H*)-quinolinone (carbostyril-124) chromophore were presented. These ligands are very efficient energy transfer donors to both Eu(III) and Tb(III) ions [141]. Whereas 7-amino-4-trifluoromethyl-2-(1*H*)-quinolinone covalently attached to the DTPA or TTHA is an effective antenna molecule for europium, but not terbium, luminescence and the emission of Eu(III) is about three times brighter than in the case of the non-fluorinated carbostyril-124 [142]. Ligand-mediated Eu(III) luminescence in complex with 4-trifluoromethylcarbostyril derivative of DTPA was also observed and applied in optical pH sensor [143].

The highly sensitive ligand, 4,7-bis(chlorosulphophenyl)-1,10-phenanthroline-2,9-dicarboxylic acid, was proposed for the determination of Eu(III) with time-resolved spectrofluorimetry (detection limit 3×10^{-13} mol/l) and for application in fluoroimmunoassays [144] as well as for the direct determination of nucleic acids in agarose gels [145]. The enhancement of Eu(III) luminescence by the formation of complexes with derivatives of 1,10-phenanthroline-2,9-dicarboxylic acid and terpyridine-6,6″-dicarboxylic acid [146] as well as by the formation of mixed ligand complexes with above-mentioned ligands (as sensitizers) and derivatives of EDTA (as shielding ligand) was discussed [147]. The energy transfer processes in lanthanide ternary complexes with 1,10-phenanthroline and various β-diketones were described in [17,28,34,35,40–45].

2.7. Nucleic acids

The energy transfer from guanosine residues to Eu(III) and Tb(III) ions, in complexes with guanine-containing nucleotides and single-stranded oligomers, was applied in a probe of DNA structure [148]. The intramolecular energy transfer process to Tb(III) from deoxyguanylic acid was the basis of its selective determination in the presence of other nucleotides [149]. The efficiency of energy transfer from nucleic acids to Tb(III) was greatly increased in the presence of phenanthroline and this system was used to the nucleic acids determination [150].

The detection of higher-ordered sequences of DNA, such as the dyad symmetrical sequence (which plays important role in organisms), was investigated using Tb(III) luminescence, based on energy transfer from excited bases in a single-stranded DNA probe [151]. Enhanced luminescence resulting from energy transfer from nucleic acids to Tb(III) ions was utilized to investigate the binding of the ions to the bases and nucleotides, as well as in the detection of single mismatches in duplexes [152].

The luminescence of Tb(III) in complexes with nucleic acids can be increased by Gd(III) (or Lu(III)) ions. Besides the intramolecular energy transfer also the intermolecular energy transfer from the DNA bound with Gd(III) to Tb(III) was observed. This columinescence system was applied to the determination of nucleic acids [153].

2.8. Indole derivatives

The luminescence of Tb(III) in complexes with indole-derived acetylacetones was studied. It was evidenced that the suitability of the energy gap between the excited triplet level of ligand and the lowest energy level of Tb(III) was a critical factor for sensitized Tb(III) luminescence and that its high intensity should be caused by a rigid planar molecular structure of the complex [154]. The Tb(III) emission due to the energy transfer from the tryptophan excited state was observed during the spectroscopic studies on calcium depleted horseradish peroxidase and used to calculate tryptophan–heme and tryptophan–Tb(III) distances in this enzyme [155].

Indole sensitized luminescence was observed when the Yb(III) or Eu(III) ions were bound at the Ca-binding sites in codfish parvalbumin (single tryptophan-containing calcium binding protein). The authors interpreted these observations in terms of an electron transfer mechanism, wherein excited singlet state tryptophan transferred an electron to the Ln(III) ion, reducing it to the Ln(II) ion; the electron then returned to the tryptophan radical cation producing Ln(III) ion in electronically excited state. Yb(III) and Eu(III) are established as redox probes of long-range electron transfer in protein [156,157]. The tryptophan fluorescence quenching by Ln(III) ions (Eu, Dy, Nd, Tb and Yb) was investigated and it was shown that the quenching strongly depended on the nature and pH of the aqueous media used [158,159].

2.9. Other heterocyclic compounds

Sensitized emission of Tb(III) ions complexed with orotic acid (2,6-dihydroxypyrimidine-4-carboxylic acid) and reversed-phase ion-pair chromatography were used for the determination of orotates in urine [160]. Intramolecular energy transfer from bleomycins and nalidixic acid to Tb(III) allowed the fluorimetric determination of these antibiotics on separation by liquid chromatography with reversed phases [161].

The intramolecular and intermolecular energy transfer processes from ligand to lanthanide ions were observed in the Sm(III) (or Tb(III))-1,6-bis(1′-phenyl-3′-methyl-5′-pyrazolone-4′)-hexanedione Gd(III)–CTMAB columinescence system and used for the determination of Sm(III) and Tb(III) with detection limits of 2×10^{-8} and 2×10^{-12} mol/l, respectively [162,163]. The luminescence of a series of Tb(III) complexes with 1-phenyl-3-methyl-4-R-5-pyrazolone based on ligand to metal energy transfer was analyzed. It was stated, that the luminescence quantum efficiency of the Tb(III) complexes increased as the R changes from an electron acceptor to an electron donor group [164].

The energy transfer between excited bromadiolone (4-hydroxy-coumarin derivative) and Tb(III) in the presence of Triton X-100 was used for the determination of this rodenticide [165]. Energy transfer from ligand, i.e. DTPA conjugated with 7-amino-4-methyl-coumarin or 1-aminonaphthalene largely enhanced the europium luminescence [166]. Incorporation of acridone as a chromophore into the 1,4,7-tris(*tert*-butoxycarbonylmethyl)-1,4,7,10-tetraazacyclo-dodecane chelating ligand allowed observation of sensitized luminescence from the Eu(III) complex [167].

A series of azatriphenylene derivatives was presented as an efficient class of antenna chromophores for luminescent lanthanide ions. They show effective energy transfer to Eu(III) and Tb(III) [168] as well as to Yb(III), Nd(III) and Er(III) [169] in acetonitrile solution. It was shown, that the successful sensitization of the lanthanide luminescence by azatriphenylenes could be expanded to aqueous media by the addition of carboxylate groups on the triphenylene skeleton [170].

The Yb(III) porphyrins were reported to show the lanthanide luminescence upon excitation of the porphyrin and were applied in in vivo luminescence localization of tumors [171]. The luminescence of the series of Yb(III)–porphyrinate complexes, excited with visible light, was studied and it was concluded that luminescence efficiency was greatly affected by the side moiety of the porphyrin ring [172].

2.10. Supramolecular compounds (with various chromophores)

Antenna effect in encapsulation complexes of lanthanide ions and the processes involved in the conversion of UV light absorbed by the ligands into visible light emitted by the Eu(III) and Tb(III) ions were extensively described [173–176]. In these works were presented 2.2.1 cryptates, tris-bipyridine cryptates, N-oxide cryptates, complexes of branched macrocyclic ligands, podates and complexes of functionalized calixarenes, which may be treated as promising labels for fluoroimmunoassays. It was concluded that the Tb(III) complexes should be more promising emitting species than the Eu(III) complexes because of the non-radiative deactivation due to LMCT states in the Eu(III) complexes and also considering that deactivation via vibronic coupling was less efficient for the Tb(III) ion [174].

The supramolecular compounds are permanently studied in many laboratories. The efficient intramolecular energy transfer from phenyl to Tb(III) ions in complexes with tetraaza phosphinate ligand was observed [177]. The ligand tetrakis(*N*-benzylcarbamoylmethyl)-tetraazacyclododecane is a suitable one for sensitizing of Tb(III), while the Eu(III) luminescence is only poorly sensitized, probably because of photoinduced electron transfer processes [178]. Similarly, the ligand *N*-(2-phenyl-1,2-dicarbonyl)-ethyl-2,3-benzo-1,4,7,13-tetraoxa-10-azacyclopentadecane displays a good antenna effect for Eu(III) luminescence, while *N*-acetic acid-2,3-benzo-1,4,7,13-tetraoxa-10-azacyclopentadecane is especially effective in transferring energy from its triplet state to Tb(III) [179]. The efficient intramolecular sensitization of Tb(III) and Eu(III) by ligand in which a benzophenone moiety was coupled to tetraazacyclododecane-tetra-acetic acid was observed [180]. Sensitized by cryptand luminescence of the Eu(III), in a containing polyamine chain and dipyridyl cryptate complex, is strongly affected by pH of solution displaying a maximum at neutral pH and significant decreases both at acidic and alkaline pH. It is due to protonation of the polyamine chain inserted within the ligand backbone. The authors defined this complex as a “pH modulated antenna device” [181]. The efficient sensitized emission of Tb(III) was observed in complex with tris(methylene phosphonic acid *n*-butyl ester)-tetraazabicyclo-pentadeca-triene. It was shown that this polyazamacrocyclic chelate of Tb(III) could be a good luminescent marker of diseased tissue [182].

The mechanism of the luminescence of Tb(III) macrocycle(tetraoxadiazacyclooctadecane) appended to the β-cyclodextrin, sensitized by the biphenyl associated with cyclodextrin cup was studied. In this system, the indirect excitation of the terbium via energy transfer from biphenyl chromophore was observed. The role of the Tb(III) ion as not only emitting center but also enhancing intersystem crossing from singlet to triplet excited state of the donor via an external heavy atom effect was emphasized [183]. In the Tb(III) and Eu(III) complexes with monoamide β-cyclodextrin derivatives of tetraazacyclododecane tetraacetate, non-covalent binding of naphthalene chromophores also leads to sensitization of the lanthanide luminescence [184]. The correlation between energy transfer efficiency and ligand structure for Tb(III) complexes with three polyoxyethylene compounds having two fluorescent chromophores (xanthene and/or benzene moieties) at both terminals was investigated. The highest value of quantum yield of Tb(III) emission for ligand with two xanthene chromophore was found [185].

Some ligands have been presented giving with lanthanides self-assembled triple-helical complexes homodinuclear: bis{1-methyl-2-(6′-[1″-(3,5-dimethoxybenzyl)benzimidazol-2″-yl]-pyrid-2′-yl)-benzimidazol-5-yl}methane (in acetonitrile) [186] and bis{1- ethyl-2-[(6′- carboxy)- pyridin-2′-yl] benzimidazol-5-yl}methane (stable in water) [187,188] as well as heterodinuclear with Zn(II): 2{6-[*N*,*N*-diethylcarbamoyl]pyridin-2-yl}-1,1′-dimethyl-5,5′-methylene-2′-(5-methylpyridin-2-yl)bis[1*H*-benzimidazole] (in acetonitrile) [189] and 2-(6-carboxypyridin-2-yl)-1,1′-dimethyl-2′-(5-methylpyridin-2-yl)-5,5′-methylenebis(1*H*-benzimidazole (in acetonitrile and in water) [190]. In these complexes, the intramolecular energy transfer between ligand and Ln(III), mainly Eu(III) ions, takes place. In the studies of the Eu(III) and Tb(III) complexes with encapsulating polybenzimidazole tripodal ligands, substituted tris(*N*-alkylbenzimidazol-2-yl-methyl)amines [191] and unsymmetrical bis(2-benzimidazolylmethyl)-(2-pyridylmethyl)amine [192], it was ascertained that the ligand-centered excitation produces the metal-centered high-intensity luminescence. It was described that pyrazole units provide an efficient energy transfer to Eu(III) and Tb(III) excited state in complexes with 1,2,4,5-tetrakis(pyrazol-1-ylmethyl-3-carboxylic acid)-benzene podand. Addition of β-cyclodextrin to solution of 1:1 Tb(III) podate leads to a large increase in luminescence intensity at physiological pH [193].

The functionalized calixarenes are one of the classes of encapsulating ligands able to form Eu(III) and Tb(III) complexes that exhibit metal luminescence upon ligand excitation. In calix[4]arenes bearing three amide groups (metal binding site) and one sensitizer group (phenacyl or diphenacylcarbonyl) on the lower rim, the excited state energy is efficiently transferred from the sensitizer to the bound Tb(III) and Eu(III) ion [194]. The luminescence characteristic of a series of calix[4]arenes with different sensitizer chromophore (naphthalene, phenanthrene or triphenylene) attached to the lower rim was described. Especially, a triphenylene antenna enables excitation of complexed Eu(III) and Tb(III) ions with near-UV wavelengths (up to 350 nm) suitable for practical applications in bioassays [195]. Sensitized near-infrared emission from Nd(III) and Er(III) complexes of fluorescein-bearing calix[4]arenes was observed. Nd(III) luminescence (at 1060 nm) is increased up to seven times upon excitation via fluorescein relative to calix[4]arene excitation. Er(III) luminescence (at 1535 nm) could only be observed upon fluorescein excitation (i.e. in visible part of the spectrum) [4]. The luminescence properties of Nd(III), Eu(III), Tb(III) and Yb(III) complexes with phenolic-type calix[4]arenes were investigated. The highest values of relative luminescence quantum yield were observed for complexes with ligands possessing methoxy- and (methoxycarbonylmethyl)oxy substituents (the most efficient ligand to metal energy transfer) [196]. In order to increase the intensity of the europium and terbium luminescence, the 6-methyl-2,2′-bipyridine chromophores at the lower rim of the calix[4]arene 1,3-bisamide were introduced [197,198]. In the Eu(III) complex of *p*-*tert*-butylcalix[4]arene with substituted 4-(2-bromoethoxy)-2,6-bis(benzimidazolyl)pyridine the overall antenna effect of both parts of the ligand was a source of the relatively strong Eu(III) luminescence [199].

The luminescence properties of the Eu(III) and Tb(III) complexes with a new class of calix[4]arene crown ethers with one or two bipyridine chromophores appended to the polyether ring (lariat calixcrowns) were studied in acetonitrile. The values of the metal-luminescence intensities obtained are among the highest for Eu(III) and Tb(III) complexes with encapsulating ligands [200]. The complex of Tb(III) with macrobicyclic ligand bis(*N*,*N*-diethylaminecarbonyl-methoxy)-*p*-*tert*-butylcalix[4]arene-crown-5 exhibits metal luminescence upon ligand excitation [201].

In acetonitrile solution, *p*-*tert*-butylcalix[5]arene forms dimeric complexes with Ln(III) ions and sensitizes luminescence of Tb(III), while in the case of Eu(III) a total quenching of the metal luminescence by a LMCT state is observed [202]. An efficient energy transfer from the ligand to the Tb(III) occurs also in dimethylformamide solution of Tb(III) *p*-*tert*-butylcalix[8]arene complex [203].

2.11. *Miscellaneous*

The energy absorbed by the chromophore moiety (carbonyl group) at the end of the cleaved polymer chain, produced in polyacrylic acid solution by the reaction with OH radicals, is transferred to Tb(III) ions resulting in an increase of their luminescence intensity. It is suggested that this system may be used as a convenient and fast OH radical detector in an aqueous solution [204]. The luminescence properties of Ln(III) complexes with the sodium salt of styrene and maleic acid copolymer in aqueous solution were investigated. Upon excitation of the styrene subunit (benzene moieties), energy migrated from the polymer to Ln(III) ions and luminescence enhancement of Tb(III), Eu(III), Sm(III) and Dy(III) were observed [205]. It was shown that in the ternary system Eu(III)–DBM–oligomer styrene-co-acrylic acid (OSAA) the luminescence of the Eu(III) was enhanced in comparison to that in Eu(III)–OSAA system. In this ternary system, two intramolecular energy transfer processes may exist simultaneously: (1) the excitation energy is absorbed by DBM and transferred to Eu(III) ion, and (2) the excitation energy is first absorbed by OSAA, then transferred to DBM in coordination sphere, and finally to the Eu(III) ion [206]. The luminescence enhancement of Tb(III) ion bound to amphiphilic linear-dendritic block copolymers (polyacrylic acid–dendritic polyether) in aqueous solution was observed due to energy transfer from the aryl dendritic framework to the bound Tb(III). The increase of Tb(III) luminescence intensity obviously enlarges with increasing generation number of the dendritic block [207].

When the EDTA and DTPA complexes of Tb(III) or Eu(III) were excited at wavelength below 250 nm, the typical lanthanide emission due to the energy transfer from the ligand was observed. The energy transfer process is more efficient in the EDTA complexes and allows determination of Tb(III) (with detection limit of 6×10^{-7} mol/l) in water, without detergents, synergistic agents or aromatic sensitizers [208].

The intracomplex energy transfer from organic dyes to near-infrared luminescent Ln(III) ions was established in the studies of the neodymium, erbium and ytterbium complexes containing DTPA and fluorescein or eosin as sensitizing chromophores excited with visible light [209]. The sensitization efficiency is higher in the case of Nd(III), Er(III) and Yb(III) complexes with the 4′,5′-bis[*N*,*N*-bis(carboxymethyl)aminomethyl] fluorescein (fluorexon) as a result of lanthanide-enhanced intersystem crossing in the sensitizing chromophore and rapid intracomplex energy transfer [210]. The luminescent label for fluoroimmunoassays based on Yb(III) complex with the ligand structurally similar to fluorexon, but carrying an isothiocyanate group reactive towards amine groups in the proteins (2′,7′-dichloro-4′,5′-fluorexon-4-isothiocyanate) was presented [211].

The luminescence of Sm(III), Nd(III) and Yb(III) complexes with xylenol orange in micellar aqueous and dimethylsulphoxide solutions was studied. Because the triplet level of xylenol orange is higher than the excited levels of Nd(III) and Yb(III), but lower than the excited level of Sm(III), in their complexes the energy transfer from the ligand only to Nd(III) and Yb(III) is possible. It permits of selective determination of Nd(III) and Yb(III) microamounts in Sm_2O_3 [212].

The luminescence of Dy(III) (also Eu(III) and Tb(III)) is enhanced when the solutions of their complexes with glucose are heated (15 min at 175 °C). This is due to the energy transfer between the aldehyde, formed when the glucose is heated, and Ln(III) ions. This effect was used for glucose determination in aqueous solution [213].

The luminescence of Tb(III) complexes with glycyl–leucyl–phenylalanine and its homologues, formed in methanol solution, was studied. Energy transfer from ligand to Tb(III) was found to be similar for all complexes investigated, but in the case of oligopeptydes with C-terminal phenylalanine the luminescence intensity was higher than that observed for other containing N-terminal phenylalanine [214].

It was shown that the $[EuN_3]^{2+}$ complex is an acceptor of the energy transferred from the singlet oxygen dimers, and simultaneously is the main chemiluminescence emitter in the system containing the Eu(II)/Eu(III)–hydrogen peroxide–azide ions [215,216].

The use of Ln(III) sensitized luminescence, based on the intramolecular and intermolecular energy transfer processes, for the determination of various organic compounds [217] and for the detection of drugs and xenobiotics in liquid chromatography [218] was presented.

3. Lanthanide ions as energy donors

3.1. Energy transfer between lanthanide ions

The dinuclear Ln(III) chelate creates an interesting basis to study lanthanide-specific energy transfer pathways inside different chelating moieties. Energy transfer from Ce(III) (d → f transitions) to Eu(III) and Tb(III) was observed to occur in binuclear complexes of diethylenetriamine-pentakis (methylphosphonic acid), ethylenediaminetetrakis(methylphosphonic acid) and hexamethylenediaminetetrakis(methylphosphonic acid) in aqueous solution [219]. Energy transfer from Eu(III) to Nd(III) in the ternary complexes with some β-diketone ligands (4,4,4-trifluoro-1-(2-thienyl)-1,3-butanedione, 4,4,4-trifluoro-1-phenyl-1,3-butanedione, 1,3-diphenyl-1,3-propanedione) and 1,10-phenanthroline shows that they are multinuclear with respect to the metal ion [220]. Intermetal ion energy transfer between Eu(III) ion and other coordinated Ln(III) ions (Nd, Pr, Ho and Er) was observed in dinuclear complexes of 1,4,7-tris(carboxymethyl)-9,14-dioxo-1,4,7,10,13-pentaazacyclopentadecane and 1,4-bis (4,7,10-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecan-1-yl)piperazine and was used to obtain structural information about these complexes [221].

In heterodinuclear lanthanide complexes with triple-helical ligand, the energy transfer from Tb(III) to Eu(III) was explained in terms of dipole–dipole mechanism [188]. Energy transfer rate constants between two trivalent lanthanide ions was determined at a short and fixed distance of ca. 4 Å, by using mixed dimers of tris(acetylacetonato)–Ln(III) complex formed in chloroform. The Ln(III) ions used as energy donors and/or energy acceptors were Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er and Yb. It was proposed that, for energy transfer at short distances, the electron-exchange mechanism was predominant over the dipole–dipole or dipole–quadrupole interactions [222]. The inter-Ln(III) energy interactions in self-assembled heterodinuclear Eu(III)–Ln(III) (Ln(III) = Er, Ho, Nd, Pr, Yb, Y) [223] and Tb(III)–Ln(III) (Ln(III) = Dy, Nd, Yb, Y) [224] chelates of 2,6-bis[*N*,*N*-bis(carboxymethyl)-aminomethyl]-4-benzoylphenol] were studied and the pathways of these interactions depending on the Ln(III) cation discussed.

The mechanisms of non-radiative transitions in trivalent lanthanide ions in solutions were discussed and it was shown that the stability constants could be determined and labile bridge binuclear complexes of Ln(III) ions may be detected using energy transfer between lanthanide ions [225]. The influence of anions on energy transfer from the Tb(III) ion to Nd(III), Sm(III), Eu(III), Er(III) and Yb(III) ions was studied. On the basis of the dependence of the quenching of Tb(III) luminescence by Nd(III) ions on the concentration of F^- anions, the method of determination of F^- ions in water was proposed [226].

Intermolecular energy transfer between Tb(III) (donor) and Eu(III) (acceptor) complexes of triethylenetetraaminehexaacetic acid was investigated and used to define the complex structure in solution [227]. The mechanism of energy transfer from Ce(III) to Ce(IV) ions in hydrogen peroxide induced Ce(III)–Ce(IV) mixture solution was studied in [228].

Intermolecular energy transfer from the excited $Tb(DPA)_3{}^{3-}$ ion to $Ln(DPA)_3{}^{3-}$ (Ln = Pr, Nd, Sm, Eu, Ho, Er) on the surface of the cetyltrimethylammonium (CTA) micelles was observed because the association of $Ln(DPA)_3{}^{3-}$ anions with the CTA micellar surface brings the lanthanide complex anion close enough to each other, so that energy transfer could occur (by an electric-dipole mechanism) [229]. Energy transfer from single molecule of $[Tb(acac)_3Phen]$ solubilized within SDS micelles to $Ln(III)_{aq}$ ions (Pr, Nd, Er and Ho) associated with the surface of the micelle was also demonstrated [230].

3.2. Energy transfer from lanthanide ions to other species

Both intra- and intermolecular energy transfer processes from lanthanide ions to other species were observed. Competition between the mechanisms of intermolecular and intramolecular energy transfer from excited Eu(III) (as $Eu(fod)_3$) to adamantanone in toluene was described [231].

Luminescence of Eu(III) and Tb(III) in methanolic solutions in the presence of some d-transition metal complexes with *N*,*N*′-bis(salicylidene)ethylenediamine, *N*,*N*′-bis(salicylidene)-1,3-propanediamine and *N*,*N*’-bis(1-methyl-3-oxobutylidene)ethylenediamine was effectively quenched by the energy transfer from the excited Eu(III) or Tb(III) to the Cu(II), Ni(II) or Cr(III) center, through the bridging oxygen atoms in formed heteronuclear complex [232]. The luminescence of Ce(III) aqua ion was quenched after binding the Ce(III) ions with tRNA and this phenomenon was used to investigation the binding of Ce(III) ion with tRNA [233].

In the system consisting of Tb(III) (or Eu(III)) ions, 2,2′-bipyridine and poly(ethylene glycol) besides strong lanthanide emission due to ligand to metal energy transfer also a bright broad blue luminescence of Ln(III)–bipyridine complex was observed. This emission is most probably due to the metal to ligand intracomplex charge-transfer [122]. It is worth to note also the investigation of the intermolecular energy transfer between phenanthrene and lanthanide ions in aqueous micellar solution. Energy transfer occurs from the lowest excited triplet state of phenanthrene (solubilized within CTA micelles) to the f^n states of Eu(III), Dy(III) and Tb(III) DPA complexes on the surface of the micelles. For the Tb(III) complex, the energy transfer is observed also from the $^5D_4$ excited state to the coexcited phenanthrene lowest triplet, thereby exciting the phenanthrene to a higher triplet state [234].

The transfer of electronic excitation energy from small Tb(III) complexes with different charges (ethylenediaminetetraacetate, (hydroxyethyl)ethylenediaminetriacetate and bis(hydroxyethyl)ethylenediaminediacetate) to chromophores in solution was employed in studies of chromophores in proteins and DNA-binding drugs [235]. The energy transfer from the excited Ce(III) ions, formed in the chemiluminescent reaction oxidation of drugs containing a thiol-group by Ce(IV) ions, to the fluorescent dye present in solution was used for the determination of these drugs [236,237].

The europium terpyridyl chelate system was used as the donor and the natural allophycocyanin dye as the energy acceptor and described as useful for delayed fluorescence resonance energy transfer (DEFRET) assay [130]. The luminescence of the Eu(III)–terpyridine–polyaminopolycarboxylate chelate was efficiently quenched by aqueous Cu(II) ions and this phenomenon was used in assay for copper at trace level [238]. Quenching of the europium terpyridine chelate emission by the base form of bromothymol blue was used for the optical measurement of pH [239].

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