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Review

A Review on the Potentiometric Lanthanum Sensors

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Abstract- Given the importance of La^{3+} ions in various areas of medicine and industry and the effects these ions can have on humans and animals, the analysis of the species is an important area in analytical chemistry. In the light of the efficiency of the ion-selective electrodes in the determinations of traces amounts of various analytes, including rare earth ions and lanthanum, all published reports on the development of La^{3+} ions were reviewed with a focus on the ionophores incorporated therein, their composition and the response and selectivity behavior of the developed sensors, to provide the readers with insight on the various factors influencing the analytical performance of lanthanum selective sensors ions, which can also be applied to research on the development of other rare earth ion sensors.

Keywords- Potentiometry; Lanthanum; Sensor; Ionophore; Ion selective electrode

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1. INTRODUCTION

La_2O_3 is used in the composition of optical glass, in hydrocarbon cracking catalysts, in carbon arcs, and in removing sulfur, and carbon in steel industries [1]. $LaCl_3$, on the other hand, has anti-tumor effects [2]. There are also reports on the application of lanthanum nitrate in enhancing the germination of rice seeds, which led to improved rate and index of germination, and vigor index. The presence of lanthanum nitrate has been reported to lower the permeability of the plasma membrane. This happens since the amounts of superoxide and malondialdehyde are lowered given that in the presence of lanthanum ions superoxide dismutase, catalase and peroxidase show higher respiratory rates and activities [3].

Lanthanum is also widely used in batteries [4], ion-selective electrodes, and chemical sensors [5,6]. La^{3+} is known to catalyze the hydrolysis of phosphate ester, which may damage the phosphate diester in DNA. This places the ions among highly toxic ions for micro and higher organisms. $LaCl_3$ possesses antitumor properties and is known to influence the peroxidation of lipids, or the activity of ATPase in the plasma membranes of rice roots. La^{3+} has been also associated with genotoxic effects on the peripheral blood lymphocytes of humans. [7,8].

Hence, in light of the considerable applications of lanthanum compounds, the chances of the occurrence of soluble lanthanum species in the environment have increased which highlights the importance of analysis of the concentration of lanthanum ions in different samples.

Commonly, trace levels of rare-earth ions are determined through various costly and time-intensive spectrophotometric techniques [9], such as ICP-AES [10], isotope dilution mass spectrometry [11], neutron activation analysis [12], and X-ray fluorescence spectrometry [13]. As an alternative potentiometric, sensors enjoy various advantages in terms of cost and ease of application, further to good sensitivity and selectivity.

Although numerous ionophores have been devised for different metal ions [14-28], the number of ionophores reported for preparing lanthanum ion sensors is rather limited. Many of the sensors prepared using the carriers have disadvantages of high limits of detection, a narrow linear response window, and considerable interference from different interfering species like Cu^{2+}, Ni^{2+}, and Ce^{3+} ions.

2. LANTHANUM SELECTIVE SENSORS

The sensors reported for the detection and quantification of lanthanum ions can be classified as polymeric membranes, carbon paste, coated graphite, and coated wire electrodes. The following lines tend to provide an overview of the various sensors prepared for lanthanum ions to date with a focus on their nature and ingredients [29-41].

The first report on a lanthanum sensor dates back to 1989 and the work of Jaber [29] who evaluated the sensing ability of PVC membranes containing tetraphenylborate salts of La complexes of nonylphenoxypolyethyleneoxyethanol, and polypropylene glycol-425. The membranes were based on 2-nitrophenyl phenyl ether or dioctylphenyl phosphonate or their mixtures as plasticizers. The membranes were reported to possess a Nernstian response (18-20 mV per decade between) in lanthanum ion solutions over a concentration range of 10^{-5} and 10^{-1} M. The optimal selectivity against the strongest interfering ions (i.e. K^+, Ba^{2+}, Pb^{2+}, and Al^{3+}) was observed in a composition of lanthanum (Antarox CO-880)TPB/ pure 2-nitrophenyl phenyl ether and tetraphenyl borate.

In 2002, Shamsipur et al developed one plasticized polymeric membrane electrode (PPME), and one membrane-coated graphite electrode (MCGE) using a cyclic ionophore with sulfur donor atoms, namely 1,3,5-trithiacyclohexane (Figure 1). The optimal membrane composition was reported to be 30% of polyvinyl chloride powder, 53% of acetophenone as the plasticizer, 2% of the ionophore and 15% of oleic acid as an ionic additive [30].

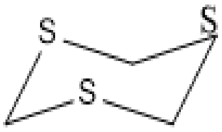

Figure 1. 1,3,5-trithiacyclohexane

The PPME had Nernstian behavior in the concentration window of 8.0×10^{-6} to 5.0×10^{-2} M, while MCGE had a wider linear response from 4.0×10^{-8} to 1.0×10^{-2} M for MCGE).

In another work, Gupta et al developed a polymeric membrane sensor for La^{3+} using monoaza-12-crown-4 (Figure 2) as the ion carrier. The optimal composition was determined to be 7% of the ionophore, 48% of PVC powder, 8% of sodium tetra phenyl borate (NaTPB) and 37% of dibutyl phthalate (DBP), which had a Nernstian response of 20.5±1.0mV from 3.16×10^{-5} to 1.00×10^{-1} M of La^{3+} concentration [31]. The maximum interference was observed for sodium ions (with a match potential selectivity coefficient of 4.2×10^{-2}), and the rest of the evaluated ions (including some common alkaline and alkaline earth and transition metal ions, and even Ce^{3+} and Eu^{3+}) caused lower interference.

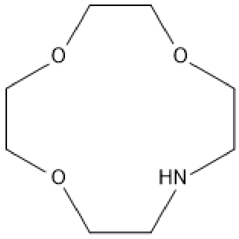

Figure 2. Monoaza-12-crown-4

In 2003 S. Khalil developed a PVC membrane sensor containing 2% of 5,14-N,N′-hydroxyphenyl-4,15-dioxo-1,5,14,18-tetraaza hexacosane (Figure 3), 10% of PVC powder, 1%

of sodium tetraphenyl borate (STPB), and 7% of dibutyl phthalate (DBP). The developed sensor had a linear calibration curve with a slope of 20 mV per decade of activity from 1.77× 10^{-6}-10^{-1} M [32].

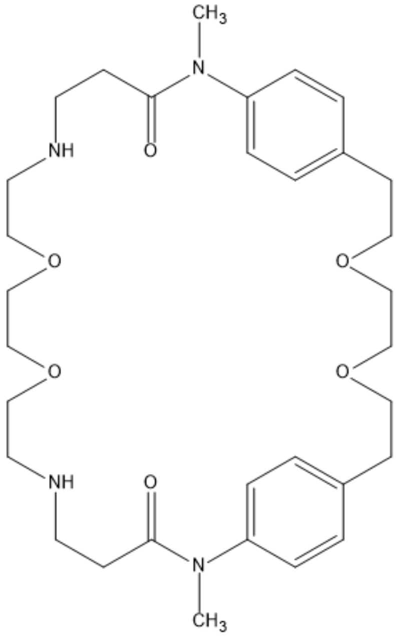

Figure 3. 5,14-N,N′-hydroxyphenyl-4,15-dioxo-1,5,14,18-tetraaza hexacosane

Ganjali et al reported a La^{3+}-selective polymeric membrane sensor (PMS) and a coated graphite membrane (CGM) using bis (2-mercaptoanil) diacetyl (Figure 4) as an ion carrier.

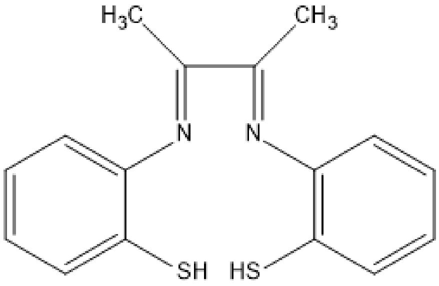

Figure 4. bis (2-mercaptoanil) diacetyl

Bis (2-mercaptoanil) diacetyl was the first semi-circular ionophore used for the development of a La^{3+}-selective sensor. The PMS and CGM had linear responses in the respective concentration widows of 1.0×10^{-5}-1.0×10^{-1} M and 1.0×10^{-6}-1.0×10^{-1} M. The maximum interference coefficients for the PMS and CGM were 3.0×10^{-2} and 7.0×10^{-3} respectively indicating the good selectivity of the sensors based on the semi-circular BMDA [33].

In another work, dicyclohexano-18-crown-6 (Figure 5) was incorporated into a membrane composition for developing a La^{3+} sensor. The optimal sensor contained 6% wt. of DC18C6, 33% wt. of PVC powder, and 61% wt. of ortho-nitrophenyl octyl ether (*o*-NPOE), and had a slope of 19mV per decade of concentration from 10^{-6} to 10^{-1} M. The maximum interferences, as determined through the matched potential and fix ion methods were from Nd^{3+} and Pr^{3+} [34].

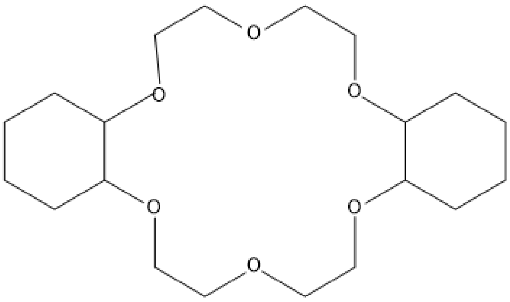

Figure 5. Dicyclohexano-18-crown-6

In 2004 Ganjali et al developed another PVC-based La^{3+} membrane electrode using bis(thiophenal)phenylen-1,3-diamine (Figure 6). This was the second lanthanum ion sensor based on a semi-circular ligand. The optimal composition of the membrane was 35% PVC, 54% of benzyl acetate, 8% of bis(thiophenal)phenylen-1,3-diamine (TPD) and 3% of sodium tetra phenyl borate and it had a response slope of 19.6 mV per decade of concentration from 1.0×10^{-7} to 1.0×10^{-1}M and a limit of detection around 3.0 ng/ml. The highest interference values were observed in the case of Sm^{3+} and Ce^{3+} ions with matched potential selectivity coefficients of 6.31×10^{-2}, and 1.99×10^{-2} reflecting the good selectivity of the sensor for analytical applications in the presence of these and other less effective interfering ions [35].

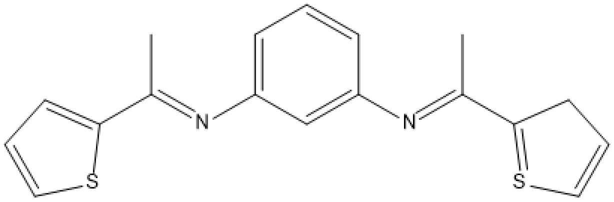

Figure 6. Bis(thiophenal)phenylen-1,3-diamine

The concept of using semicircular ligands was further verified by the results in another report on the application of an azao-containing Schiff's base (N,N′-adipylbis(5-phenylazo salicylaldehyde hydrazone (Figure 7)) for developing a La^{3+}-selective PVC membrane sensor. The optimal electrode composition was described as containing 27% wt. of PVC, 54% wt. of 2-nitrophenyl octyl ether (NPOE), 3% wt. of the ionophore, and 16% wt. of oleic acid as an ionic additive. Membrane sensors with this composition produced a linear calibration curve with a slope of 19.4 mV decade of La^{3+} concentration from 1.0×10^{-6} to 1.0×10^{-2} M and a detection limit of 0.05 ppm was also reported for the sensors. The highest interferences were reported for Al^{3+} and Cu^{2+} ions with respective pK_{sel} values of 3.22 and 3.3 [36].

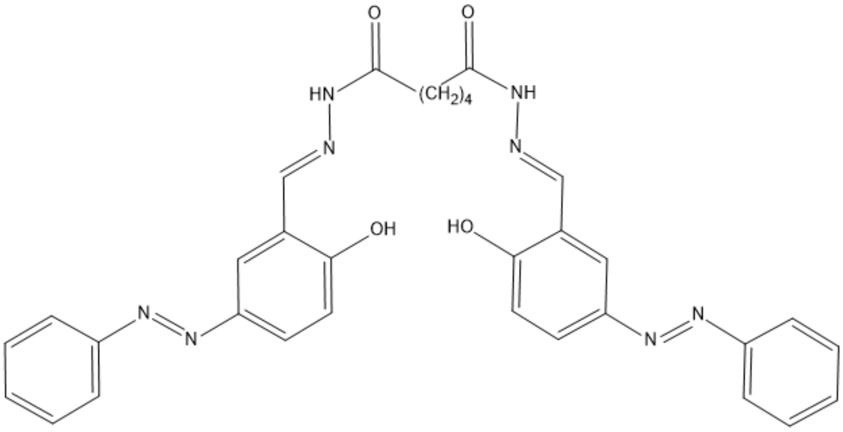

Figure 7. N,N′- adipylbis(5-phenylazo salicylaldehyde hydrazone

The trend of using open-ring ligands for developing ion-selective electrodes for La^{3+} continued with the work of Ganjali et al [37] who used amitraz (N-2,4-dimethylphenyl- N′-ethylformamidine (Figure 8)) in a PVC-based liquid membrane, with an optimal composition of 8% of amitraz, 58% of benzyl acetate, 4% of sodium tetraphenyl borate (NaTPB), and 30% of PVC powder [37]. The electrode had a Nernstian response in the range of 1.0×10^{-7} to 1.0×10^{-1} M (19.8±0.2 mV per decade of concentration). The electrode was highly selective and even the strongest interfering ion i.e. Ce^{3+}, had a selectivity coefficient of 1.10×10^{-5}.

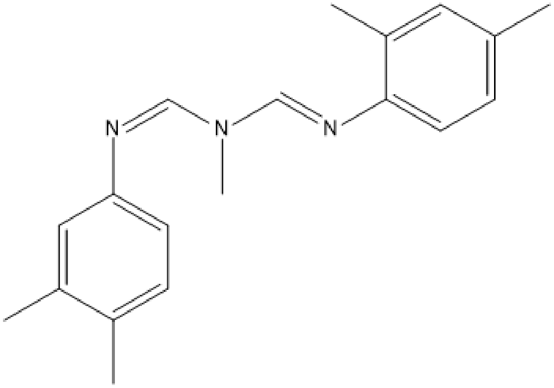

Figure 8. N-2,4-dimethylphenyl- N′-ethylformamidine

Zamani et al, reported a La^{3+}-selective membrane electrode using N′-(1-pyridin-2-ylmethylene)-2-furohydrazide (Figure 9) and observed a linear behavior with a slope of 19.2± 0.6 mV per decade from 1.0×10^{-6}-1.0×10^{-1} M. The optimal electrode composition was 30% of PVC, 62% of nitrobenzene, 5% ion-carrier, and 3% sodium tetra phenyl borate. Among the interfering ions tested (i.e. Na^{+}, K^{+}, Mg^{2+}, Ca^{2+}, Sr^{2+}, Ba^{2+}, Cu^{2+}, Co^{2+}, Zn^{2+}, Ni^{2+}, Pb^{2+}, Cd^{2+},

Ce^{3+}, Gd^{3+}, Sm^{3+}, Eu^{3+}, Nd^{3+}, Dy^{3+}) the highest selectivity coefficients were 3.4×10^{-3} and 1.7×10^{-3} for Sm^{3+} and Dy^{3+}) [38].

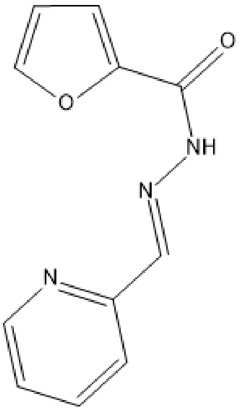

Figure 9. N′-(1-pyridin-2-ylmethylene)-2-furohydrazide

Application of cyclic ligands, on the other hand, was not completely forgotten and Singh et al. developed a La^{3+}-selective PVC membrane sensor using hexaaza macrocycle, 8,16-dimethyl-6,14-diphenyl-2,3,4:10,11,12-dipyridine-1,3,5,9,11,13-hexaaza cyclohexadeca-3,5,8,11,13,16-hexaene (Figure 10) as well as dibutylbutyl phosphonate (DBBP) as membrane solvent NaTPB, and PVC, as an ionic additive. The membrane with the optimal composition (i.e. 10:260:5:120 (I:DBBP:NaTPB:PVC)) produced a linear response with a slope of 19.8±0.2 mV/decade of concentration from 7.9×10^{-7}–1.0×10^{-1} M). The limit of detection was determined to be 5.62×10^{-7} M [39].

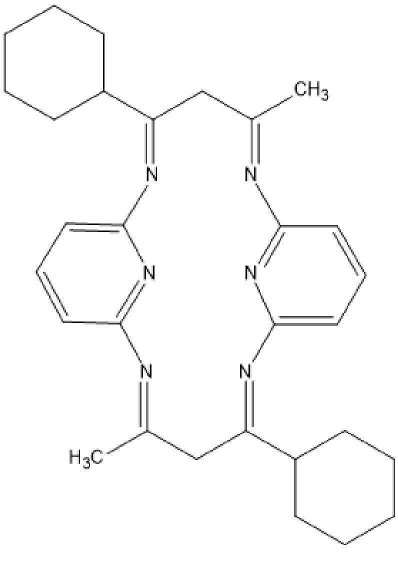

Figure 10. 8,16-dimethyl-6,14-diphenyl-2,3,4:10,11,12-dipyridine- 1,3,5,9,11,13-hexaazacyclohexadeca-3,5,8,11,13,16-hexaene

Ganjali et al., developed a La^{3+} selective electrode 3-hydroxy-N′-(pyridin-2- ylmethylene)-2-naphthohydrazide (Figure 11), further containing, potassium tetrakis (p-chlorophenyl) borate (KTpClPB) and ortho nitrophenyloctyl ether (NPOE) at values of 6%, 2%, and 62% wt. respectively in a PVC matrix. The sensor had a linear calibration curve with a slope of 19.2 mV per decade from 1.0×10^{-7}-1.0×10^{-2} M with a detection limit of about 10 ng/mL. The highest interference effects were observed in the case of Cs^{+}, Yb^{3+}, Ce^{3+} and Sm^{3+} with respective selectivity coefficients of 4.75×10^{-3}, 3.80×10^{-3}, 3.41×10^{-3}, 3.42×10^{-4} [40].

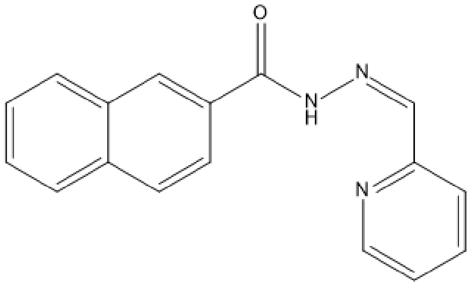

Figure 11. 3-hydroxy-N′-(pyridin-2- ylmethylene)-2-naphthohydrazide

In the same year, Ganjali et al developed a La^{3+}-selective PVC-based membrane electrode using 6% wt. of 4-methyl-2-hydrazinobenzothiazole (Figure 12), 3% wt. of NaTPB, 61% wt. of dibutyl phthalate and PVC (30%) and reported a linear response of 19.8 mV per decade of concentration from 1.0×10^{-7} to 1.0×10^{-1}. The limit of detection was evaluated to be around 3.8 ppb [41].

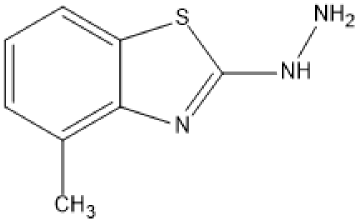

Figure 12. 4-methyl-2-hydrazinobenzothiazole

In the same year, the same author and his team reported another La^{3+}-selective electrode based on 8-amino-N-(2-hydroxybenzylidene) naphthylamine (Figure 13) as an ionophore with an optimal composition of 7% of the ionophore, 30% of PVC, 61% of NPOE and 2% of KTpClPB. (Slope: 20.3±0.3 mV per decade, from 1.0×10^{-7} to 1.0×10^{-1} M; detection limit: 8.0×10^{-8} M). They reported the highest interferences to be from Pr^{3+}, Yb^{3+}, Ce^{3+}, and Gd^{3+} with respective log pK_{sel} values of 2.6, 3.2, 3.6, and 3.9 [42].

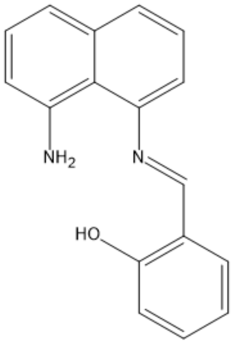

Figure 13. 8-amino-N-(2-hydroxybenzylidene)naphthylamine

Another report is on a membrane sensor for lanthanum ion using bis(5-nitro-2-furaldehyde)butane-2,3-dihydrazone (Figure 14). The composition of the optimal electrode was described as 30% (PVC), 56% (benzyl acetate), 8% (ionophore), and 6% oleic acid and a linear response (19.8±0.2 mV per decade of concentration) was recorded from 1.0×10^{-7} to 1.0×10^{-1} M (limit of detection: 5.6 ng/ml) [43].

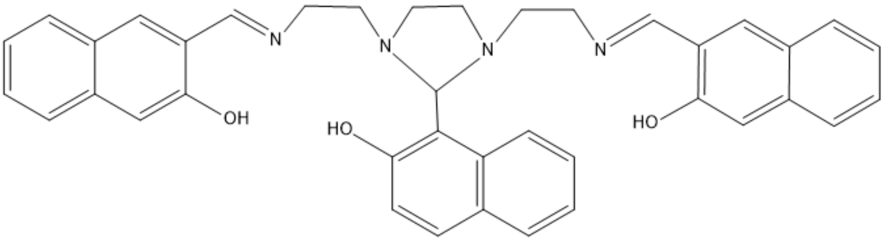

Figure 14. Bis(5-nitro-2-furaldehyde)butane-2,3-dihydrazone

In 2011, Akbar Islamnezhad et al used ethyl1,2,3,4-tetrahydro-6-methyl-4-phenyl-2-thioxopyrimidine-5-carboxylate (Figure 15) as a La^{3+} selective ion-carrier in a membrane with an optimal calibration slope of 19.9±0.3 mV/decade of concentration from 9.3×10^{-8} to 1.0×10^{-1} M. The optimal membrane composition was reported to be PVC (30% wt.): acetophenone (60% wt.): oleic acid (5% wt.) and 5% wt. of the ligand. The maximum interfering effects among the various ions evaluated were caused by Pr^{3+} (K_{sel}: 2.3×10^{-2}), and Sm^{3+} (K_{sel} :3.6×10^{-3}) indicating the good selectivity of the developed sensor [44].

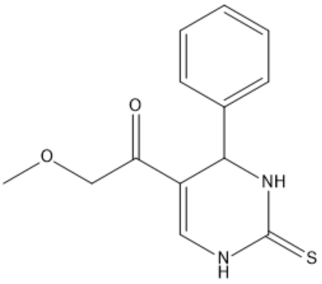

Figure 15. Ethyl1,2,3,4-tetrahydro-6-methyl-4-phenyl-2-thioxopyrimidine-5-carboxylate

S. Chandra et al, in the same year, used a cyclic ionophore i.e. 1, 12diaza-5,8,13-trioxo-3(4), 9(10)-dibenzocyclotridecane (Figure 16) in a PVC membrane further containing 52% of acetophenone, 6% oleic acid, and 35% PVC and recorded a linear response (19.4±0.4 mV/decade of concentration) in the concentration window of 3.4×10^{-7} to 1.0×10^{-2} [45].

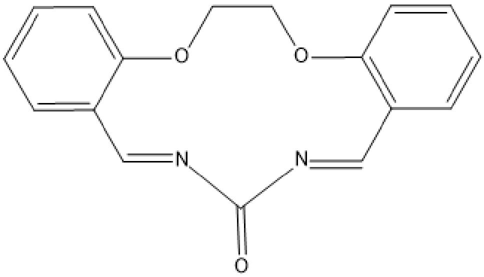

Figure 16. 1, 12diaza-5,8,13-trioxo-3(4), 9(10)-dibenzocyclotridecane

In another study, a novel La^{3+} selective graphite disk electrode was modified using a polymeric membrane containing 2.5 mg of benzo-15-crown-5 (Figure 17), 7 mg of oleic acid, 60.5 mg of *o*-NPOE and 30 mg of PVC and observed a linear response (19.5 mV per decade of concentration) from 5×10^{-7} to 1×10^{-2} M [46]. The highest interferences were caused by Cr^{3+} (K_{sel}: 2.5×10^{-3}) and Ca^{2+} (K_{sel}: 7.0×10^{-3}) [46].

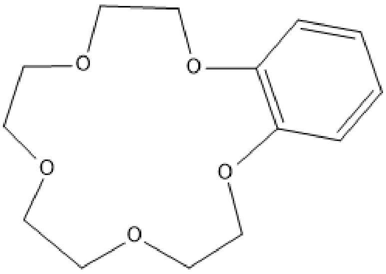

Figure 17. Benzo-15-crown-5

In 2013, two polymeric membrane and coated graphite electrodes (PME, GCE) for La^{3+} ions were developed using an acyclic ionophore, i.e. 3-amino-2-mercapto-3H-quinazolin-4-one (Figure 18) The PME had a slope of 20.1±0.3 mV/decade of concentration from 3.0×10^{-7} to 1.0×10^{-1} M, while these values were 23.4±0.4 mV/decade of concentration and 1.0×10^{-7} to 1.0×10^{-1} M. Both membranes had a composition of 31% PVC, 63% NPOE, 5% of the ionophore and 1% KTpClPB. The highest interferences in the case of the PME were caused by Ce^{3+} and Ag^{+} (K_{sel}: 2.8×10^{-2}, 8.7×10^{-2}). Among all tested interfering ions Ce^{3+}, Y^{3+}, Hg^{2+}, $K^{+,}$ and Ag^{+} had the highest interferences for the GCE response [47].

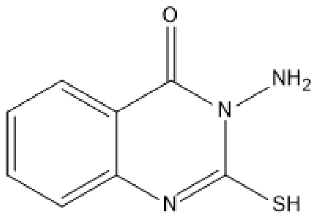

Figure 18. 3-amino-2-mercapto-3H-quinazolin-4-one

In another report researchers self-assembled 3-amino-5-mercapto-1,2,4-triazole (Figure 19) on Ag nanoparticles to prepare a self-assembled ionophore for a screen-printed electrode for La^{3+} ions. The nanoparticle screen printed electrode had a calibration slope of 20.2±0.1 mV decade-1 of lanthanum concentration, in the range of 1.0×10^{-8}-1.0×10^{-1}. It was reported that the selectivity coefficients were 8.5×10^{-2} or less, which reflects the acceptable selectivity of the sensor [48].

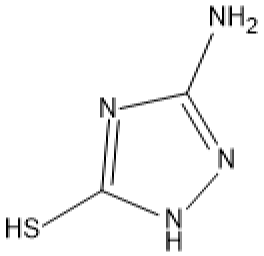

Figure 19. 3-amino-5-mercapto-1,2,4-triazole

In 2013 there was a report on the electropolymerization of pyrrole in a medium also containing benzo-15-crown-5 (Figure 17), which is a complexing ligand, to prepare a modified polymer for the development of a sensor for La^{3+} ions and a slope of 19.5 mV $decade^{-1}$ was reported in a rather narrow window of 1×10^{-4} to 1×10^{-1} M [49].

In 2016, E. Frag et al, developed a modified carbon paste and a screen-printed electrode (CPE and SPE) using ethyl benzoyl acetate (EBA) for the determination of La^{3+} ions and evaluated different plasticizers (namely onitrophenyl octyl ether, tricresylphosphate, dibutylphthalate, dioctylsebacate, and Dioctylphthalate, as well as the effect of the ionophore, and declared that the best sensor behavior for both SPE and CPE was observed when -NPOE and TCP were used in the membrane composition. Under these conditions, the respective calibration slopes of the developed SPE and CPE were 21.0, 19.5, and 20.5 mV/decade of concentration in the range of 1.0×10^{-6} to 1.0×10^{-2} M [50].

Rather recently a La^{3+}-sensor was based on impregnating a polytetrafluoroethylene membrane with tributyl phosphate (TBP) [51]. The latest work in the area is on the development of a potentiometric La^{3+} carbon paste electrode (CPEs) composed of graphite powder, an 1 -Butyl-3-methylimidazolium hexafluorophosphate, graphene oxide and 2-((2-(5-amino-1, 3, 4-thiadiazol-2-yl)hydrazono)methyl)phenol (Figure 20). A linear Nernstian behavior was reported from 7.0×10^{-7} to 1.0×10^{-1}, M and the limit of detection was reported to be 5.16×10^{-8} M [52].

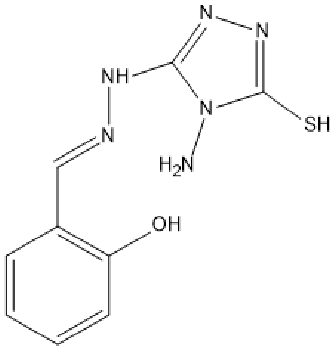

Figure 20. 2-((2-(5-amino-1, 3, 4-thiadiazol-2-yl)hydrazono)methyl)phenol

3. CONCLUSION

The majority of lanthanum selective sensors with cyclic ionophores possessed hard donor atoms like oxygen, or at least a large number of their donor atoms were hard bases, although medium N atoms are also present in some cases. Examples include monoaza-12-crown-4 [31]; 5,14-N,N′-hydroxyphenyl-4,15-dioxo-1,5,14,18-tetraaza hexacosane [32]; dicyclohexano-18-crown-6 [34]; 1, 12diaza-5,8,13-trioxo-3(4), 9(10)-dibenzocyclotridecane [45]; benzo-15-crown-5 [46]. Only in the case of 1,3,5-trithiacyclohexane [30]; and 8,16-dimethyl-6,14-diphenyl-2,3,4:10,11,12-dipyridine- 1,3,5,9,11,13-hexaazacyclohexadeca-3,5,8,11,13,16-hexaene [39] soft and medium donor atoms are present in the cyclic ionophores.

In the acyclic ionophores, the share of medium (N) and soft (S) donor atoms of the ionophore is higher (e.g. bis (2-mercaptoanil) diacetyl [33]; bis(thiophenal)phenylen-1,3-diamine [35]; 2,4-dimethylphenyl- N′-ethylformamidine [37]; 3-hydroxy-N′-(pyridin-2-ylmethylene)-2-naphthohydrazide [40]; 4-methyl-2-hydrazinobenzothiazole [41]; 3-amino-2-mercapto-3H-quinazolin-4-one [47]; 3-amino-5-mercapto-1,2,4-triazole [48]; 2-((2-(5-amino-1, 3, 4-thiadiazol-2-yl)hydrazono)methyl)phenol [52]. It is also noteworthy that O atoms are also present in considerable numbers in some acyclic ionophores, like N,N′- adipylbis(5-phenylazo salicylaldehyde hydrazine [36]; N′-(1-pyridin-2-ylmethylene)-2-furohydrazide [38]; bis(5-nitro-2-furaldehyde)butane-2,3-dihydrazone [43]; and ethyl1,2,3,4-tetrahydro-6-methyl-4-phenyl-2-thioxopyrimidine-5-carboxylate [44], but even in those cases a fair share of medium (N) or soft (S) donors are also present.

One could conclude that in the absence of rigid conformations (closed rings), where size is not as important in the selective interaction of the ionophore and the target ion and more flexible ligands have the capability of forming wrap-around complexes interact with La^{3+} ions, medium and soft donor atoms showed favorable interactions with La^{3+} ions.

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