

Review

A Review on the Development of Gadolinium Potentiometric Sensors

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Abstract- Gadolinium is a rare earth element with various applications, which has led to the distribution of water-soluble compounds of this element and has subsequently, increased the chances of exposure of humans and animals to Gd^{3+} . Given the effects this can have on human and animal health the analysis of gadolinium compounds has become more important and ion-selective electrodes constitute a set of versatile tools for this purpose. The manuscript tends to provide an overview on the elective polymeric membrane electrodes for the analysis of Gd^{3+} and their performance.

Keywords- Gadolinium; Ionophore; Ion selective electrode; Potentiometric sensor

1. INTRODUCTION

Gadolinium is a silvery, soft, and ductile lanthanide, which does not tarnish in the absence of water but is covered with a layer of its oxide in moist atmospheres. Gadolinium acts as a superconductor below 810 °C and is highly magnetic under ambient conditions. The element is used in control rods of nuclear reactors; and in the composition of garnets used in microwave devices. The element is also present in magnetic alloys or those used in electronic components like recording heads of video recorders, compact disks, and digital memories [1]. Gadolinium compounds were used in phosphorous in color televisions [2].

Like other lanthanides, compounds of gadolinium are widely used in glass formulations, glass fibers, oil and gas catalysts, polishing materials, carbon arcs, and for purification

purposes in the steel industry [1]. Compounds of gadolinium have also been used as contrast agents in magnetic resonance imaging (MRI) applications [3].

The widespread applications of various water-soluble lanthanide compounds, which have increased the risks of direct and indirect exposure to such materials in humans and animals have become a great concern [4-7]. Gadolinium can be very dangerous if its compounds are inhaled, which can lead to lung embolisms over time. Soluble gadolinium compounds can cause serious conditions since their ions bioaccumulate in the liver. In water animals, Gd^{3+} ions have been reported to damage cell membranes, adversely influencing reproductive and nervous systems.

Consequently, various analytical techniques have been reported for the determination of Gd^{3+} concentrations in different systems. These range from spectrometric techniques like spectrometry and ICP-MS [8–11] and electron spin resonance [12–14], laser-based multi-step resonance ionization [15], phosphorescence opto-sensing [16], time-resolved fluorimetry [17], spectrophotofluorimetric analysis [18], fluorescence [19], and nucleic methods [20].

An alternative approach for the analysis of gadolinium is the application of ion-selective electrodes based on solvent-based polymeric membranes [21-31]. These devices constitute a set of customizable devices, offering the advantages of low cost, portability, selectivity, quick analysis, and low detection limits. Given these advantages, a wide range of reports has been published on the application of ionophores for constructing selective sensors for lanthanide ions such as Yb^{3+} , La^{3+} , Eu^{3+} , Dy^{3+} , Tb^{3+} , Pr^{3+} , Lu^{3+} , and Sm^{3+} [32–45] apart from other cations and anions [46].

2. GADOLINIUM SELECTIVE ION SELECTIVE ELECTRODES

Regarding what has been said ion selective electrodes for the determination of Gd^{3+} ion sensors could be an effective strategy for the analysis of this ion in various samples. The first report on the development of a poly(vinyl chloride) (PVC) based membrane sensor for Gd^{3+} ions was published by Ganjali, M. R., et al. in 2003 [47]. The sensor was based on (2-[(3-[(2-sulfanyphenyl)imino)-1-methylbutylidene]amino]phenyl hydrosulfide (Figure 1) which is a Schiff's base with nitrogen and sulfur donating atoms.

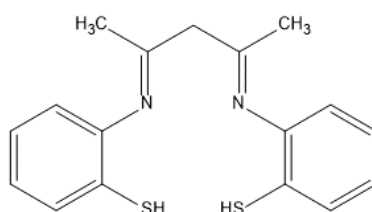


Figure 1. (2-[(3-[(2-sulfanyphenyl)imino)-1-methylbutylidene]amino]phenyl hydrosulfide

Benzyl acetate (BA), dibutyl phthalate (DBP) as the solvent mediators, and sodium tetraphenylborate (TPB) as an ionic additive were used in the electrode composition and the optimal sensor composition was reported to be 33% PVC, 61% of BA, 2% of NaTPB and 5% of the ligand, which led to a linear response with a slope of 19.8 ± 0.3 mV per decade from 1.0×10^{-5} and 1.0×10^{-1} mol L⁻¹. The response of the sensor was linear in the pH window of 4.0-8.0, and it proved to have negligible interference from Sm³⁺, Ce³⁺, La³⁺, Cu²⁺, Pb²⁺, and Hg²⁺ while the maximum interferences were reported to be caused by Eu³⁺ and Dy³⁺ with respective matched potential method selectivity coefficients (K_{MPM}) of 5.0×10^{-2} , 3.5×10^{-2} .

Almost at the same time, Ganjali, M. R., et al. reported developing another Gd³⁺ sensor based on omeprazole (Figure 2) which is an antibiotic [48]. They evaluated benzyl acetate (BA), dioctyl phthalate (DOP), dibutyl phthalate (DBP), and ortho-nitrophenyloctyl ether (NPOE) as the solvent mediators in the construction of the liquid membrane sensors and reported the optimal behavior to be observed by DMP. The best composition (i.e. 35% of PVC, 56% of DBP, 5% of the ionophore, and 4% of NaTPB) produced a linear response with a slope of 19.3 ± 0.3 mV decade⁻¹ from, the electrode has a linear dynamic range between 1.0×10^{-5} and 1.0×10^{-1} mol L⁻¹ and the response of the sensor was linear in the pH window of 4.0-10.0.

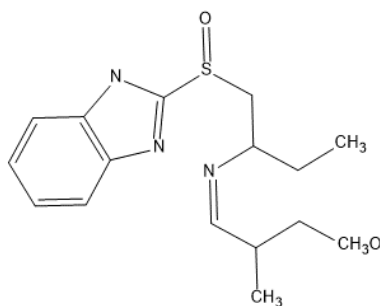


Figure 2. Omeprazole

In 2007 Zamani, H. A., et al. [49] reported a very sensitive sensor for Gd³⁺ using 6-methyl-4-[[1-(2-thienyl)methylidene]amino]3-thioxo-3,4-dihydro-1,2,4-triazin-5-(2H)-one (Figure 3), together with 32% wt. of PVC, 63% of o-NPOE as the plasticizer, and 1.5% wt. of NaTPB as an ionic additive. The other solvent mediators used (i.e. DBP and nitrobenzene could not lead to optimal results). The best sensor linearly responded to Gd³⁺ ion concentration with a slope of 19.8 ± 0.2 mV per decade from 1.0×10^{-6} - 1.0×10^{-1} mol L⁻¹ and had a detection limit of 5.8×10^{-7} mol L⁻¹. The pH range in which the sensor response was not influenced by the concentration of the proton ions extended from 3.2-8.7. Among the various ions tested (i.e. Zn²⁺, Co²⁺, Ni²⁺, Cd²⁺, Sm³⁺, La³⁺, Ce³⁺, Yb³⁺, Tb³⁺, Fe³⁺, Cr³⁺, Hg²⁺, Pb²⁺, Ag⁺, Na⁺, K⁺, Ca²⁺, Mg²⁺) the highest MPM selectivity coefficients were reported for Hg²⁺ ($K_{\text{MPM}} = 3.8 \times 10^{-3}$).

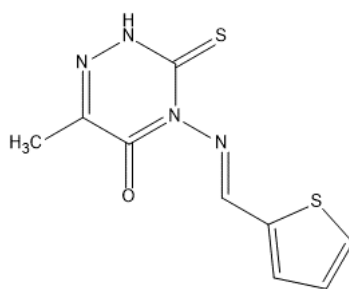


Figure 3. Structure of 6-methyl-4-[[1-(2-thienyl)methylidene]amino]3-thioxo-3,4-dihydro-1,2,4-triazin-5-(2H)-one

One year later Faridbod et al reported using N-(2-pyridyl)-N'-(4-nitrophenyl)thiourea (Figure 4) to develop an asymmetric potentiometric Gd^{3+} microsensor [50]. Like the previous ionophores used for the construction of gadolinium sensors, the acyclic ionophore used in this report also contained medium and soft donor atoms (i.e. N and S). Three solvent mediators namely DBP, nitrobenzene (NB), and BA were evaluated and the ionic additive used was potassium tetrakis (p-chlorophenyl) borate (KTPClPB).

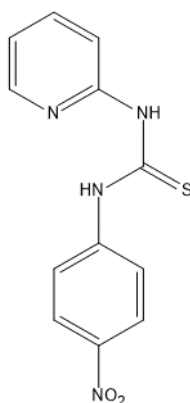


Figure 3. Structure of N-(2-Pyridyl)-N'-(4-nitrophenyl)thiourea

Based on the report the best composition included 20%wt of PVC powder, 72%wt. of BA, 5% wt of N-(2-Pyridyl)-N'-(4-nitrophenyl) thiourea, and 3% of KTPClPB led to a linear response from 1.0×10^{-8} to 1.0×10^{-3} mol L^{-1} , with a slope of 17.46 ± 0.3 mV per decade, and had a distinctly lower detection limit of 3.0×10^{-9} mol L^{-1} in comparison to the previous reports on potentiometric Gd^{3+} sensors [47-49]. The electrode response was independent of pH from 4.0 to 9.0 and the maximum interference was observed for Sm^{3+} , Eu^{3+} , Er^{3+} , Dy^{3+} with respective Log K_{MPM} values of -2.5, -2.6, -2.8, -2.9.

Sharma et al later reported a potentiometric Gd^{3+} electrode using zirconium(IV) tungstophosphate [ZrWP] as a novel ion-exchanger [51], which was in constructing an ion exchange membrane based on an epoxy resin binder with a composition of 40% of Zirconium(IV) tungstophosphate and 60% of the binder. The electrochemical membrane had a

linear response with a slope of 30 mV decade⁻¹ from 1×10^{-5} to 1×10^{-1} mol L⁻¹ with a fixed response in the pH window of 4.0-10.0 for a 10^{-1} mol L⁻¹ solution and 3.0-7.0 for a 10^{-2} mol L⁻¹ solution. The highest interference was caused by Nd³⁺ ions, which had a K_{MPM} of 5.25×10^{-2} , and the rest of the tested interfering ions (i.e. Sm³⁺, Cu²⁺, Ce³⁺, Pb²⁺, Fe³⁺, Ca²⁺, Dy³⁺, La³⁺, Pr³⁺, Na⁺, Tb³⁺) influenced the response of the electrode less.

In another work in 2009, Singh et al reported a coated graphite Gd³⁺ selective electrode using 2,6-bis-[1-{N-cyanopropyl,N-(2-methylpyridyl)}aminoethyl]pyridine (Figure 4) as an ion carrier [52].

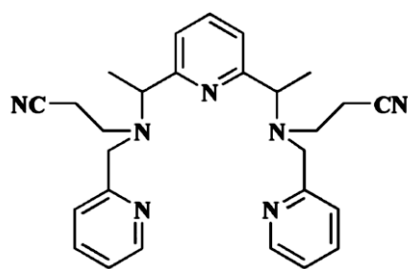


Figure 4. Structure of 2,6-bis-[1-{N-cyanopropyl,N-(2-methylpyridyl)}aminoethyl]pyridine

The film coated on the graphite element had a composition of 8% wt. of the ligand, 4% wt. of NaTPB, 30% wt. of PVC, and 58% wt. of *o*-NPOE, and led to a Nernstian response of 19.6 ± 0.1 mV decade⁻¹ from 2.8×10^{-7} to 5.0×10^{-2} mol L⁻¹ and a detection limit of 6.3×10^{-8} in the pH range of 2.0 to 8.0. A set of interfering ions (Sm³⁺, Nd³⁺, La³⁺, Ce³⁺, Pr³⁺, Tm³⁺, Tb³⁺, Cu²⁺, Zn²⁺, Cd²⁺, Co²⁺, Ni²⁺, Ca²⁺, Ag⁺, Na⁺, K⁺, Sr²⁺) were evaluated and the maximum interference was caused by Sm³⁺ with a K_{MPM} of 7.2×10^{-3} .

In 2011, Rezaei et al. evaluated 4-Methoxyphenylcyanamide as a Gd³⁺ ionophore for application in a polymeric membrane sensor [53]. Among the tested plasticizers (i.e. tributyl phosphate (TBP), DBP, BA, dioctyl phthalate (DOP), and *o*-NPOE the best response was observed with TBP and the optimal composition was reported to be 30% wt. of PVC, 60% wt. of TBP, 6.0% wt. of the ligand and 4% wt. of NaTPB. The optimal response worked well from 1.0×10^{-6} to 1.0×10^{-2} mol L⁻¹ with a slope of 19.5 mV decade⁻¹. The electrode response was independent of pH from 4.1 to 8.3 and its selectivity behavior in the presence of Na⁺, K⁺, NH₄⁺, Mg²⁺, Ca²⁺, Sr²⁺, Co²⁺, Ni²⁺, Cu²⁺, Zn²⁺, Eu³⁺, Sm³⁺, and Yb³⁺ proved its good selectivity behavior with the maximum interference caused by Eu³⁺ with a K_{MPM} value of 8.4×10^{-2} .

Zamani, et al [54] used 3-methyl-1H-1,2,4-triazole-5-thiol (Figure 5) in a membrane further composed of nitrobenzene and NaTPB to construct a membrane sensor with a linear Nernstian response of 19.8 ± 0.4 mV/decade from 1.0×10^{-7} to 1.0×10^{-2} mol L⁻¹ with a pH-independent response from 2.9-8.4.

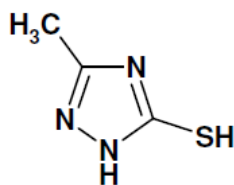


Figure 5. Structure of 4-Methoxyphenylcyanamide

Although the other plasticizers (i.e. acetophenone (AP), benzyl acetate (BA), dibutyl phthalate (DBP)) produced some sensitivity the best response was produced by nitrobenzene (NB) and the best response was reported for a membrane sensor composed of 30% wt. of PVC, 66% wt. of NB, 2% wt. of NaTPB and of 4-Methoxyphenylcyanamide each. No report was made on the selectivity coefficients of the sensor for different interfering ions.

Zamani, et al. also reported a Gd^{3+} selective sensor using N,N'-bis(methylsalicylidene)-2-aminobenzylamine (Figure 6) using different solvent mediators (NB, AP, BA, DBP) and NaTPB as the ionic additive [55]. For an optimal composition of 30% PVC, 66% AP, 2% NaTPB, and 2% N,N'-bis(methylsalicylidene)-2-aminobenzylamine they reported a response slope of 19.7 ± 0.3 mV/decade from 1.0×10^{-6} to 1.0×10^{-2} mol L^{-1} . The response of the sensor was independent of the pH of the test solution from 3.5 to 10.1. The selectivity of the sensor for Gd^{3+} was evaluated in the presence of Tm^{3+} , Er^{3+} , Lu^{3+} , Tb^{3+} , Dy^{3+} , Ho^{3+} , Cr^{3+} , K^{+} , Mg^{2+} , Ca^{2+} , Pb^{2+} , Co^{2+} , Ni^{2+} and the maximum interference was observed for Co^{2+} , Ni^{2+} with the K_{MPM} values of 5.2×10^{-3} and 4.3×10^{-3} .

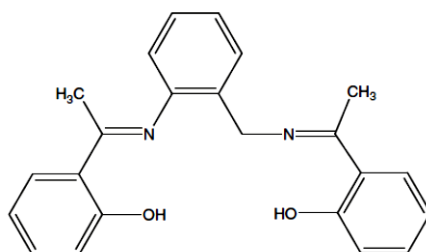


Figure 6. N,N'-bis(methylsalicylidene)-2-aminobenzylamine

Two gadolinium selective liquid membrane sensors were reported by Gupta et al [56], using two different ionophores namely 2-(2-aminothiazol-4-yl)phenol (Figure 7) and 2-(4-phenyl-1,3-thiazol-2-yl)iminomethyl)phenol (Figure 8).

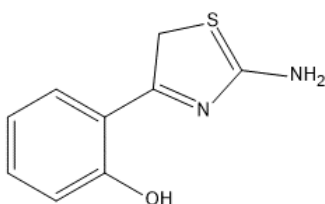


Figure 7. Structure of 2-(2-aminothiazol-4-yl)phenol

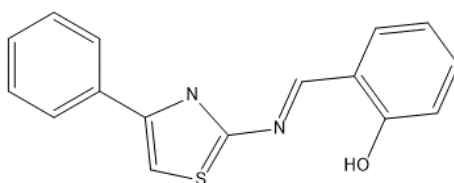


Figure 8. Structure of 2-(4-phenyl-1,3-thiazol-2-yliminomethyl)phenol

Initial evaluations revealed that 2-(4-phenyl-1,3-thiazol-2-yliminomethyl)phenol led to higher sensitivity in comparison to 2-(2-aminothiazol-4-yl)phenol and that among AP, DBP, *o*-NPOE, tributyl phthalate (TBP), 1-chloronaphthalene (CN), dioctyl phthalate (DOP) the best response was observed with AP. The membrane containing 5% 2-(4-phenyl-1,3-thiazol-2-yliminomethyl)phenol, 33% PVC, 66% of AP, and 2% NaTPB was reported to have a Nernstian slope of 19.5 ± 0.5 mV per decade from 9.4×10^{-7} to 1.0×10^{-2} mol L⁻¹ in the pH range of 3.5-8.5. Among the interfering ions evaluated (i.e. Li⁺, Na⁺, K⁺, Mg²⁺, Zn²⁺, Co²⁺, Ni²⁺, Cu²⁺, Al³⁺, Nd³⁺, Ce³⁺, La³⁺) with the sensor based on the better ionophore the highest interferences were observed for Ce³⁺ with a selectivity coefficient of 5.76×10^{-2} .

In a later work, Vahdani, M., et al. [57] reported a liquid membrane sensor using 1-(4-nitrophenyl)-3-(4-phenylazophenyl)triazene (Figure 9) with a linear response of 19.9 ± 0.2 mVdecade⁻¹ from 1.0×10^{-6} to 1.0×10^{-2} in ($2.6 < \text{pH} < 8.7$). The best composition included 30% PVC, 4% NaTPB, 60% nitrobenzene, and 6% 1-(4-nitrophenyl)-3-(4-phenylazophenyl) triazene. Oleic acid was also used as an ionic additive together with NaTPB but the response was not satisfactory. The maximum interference was observed for Ca²⁺ with a selectivity coefficient of 8.0×10^{-3} .

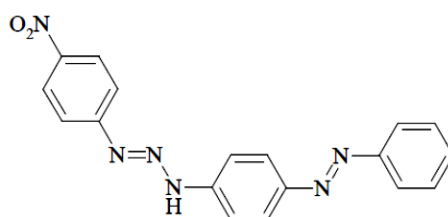


Figure 9. 1-(4-nitrophenyl)-3-(4-phenylazophenyl)triazene

N'-(2-oxo-1,2-di(pyridin-2-yl) ethylidene)furan-2-carbohydrazide (Figure 10) has also been reported as a Gd³⁺ selectophores for construction of liquid membrane sensor [58]. Various compositions containing DBP, NB, BA, *o*-NPOE, and AP as the solvent mediators and NaTPB, and oleic acid as the ionic additives revealed the optimal composition to be 30% PVC powder, 51% NPOE, 6% of the ionophore, and a mixture of the additives (i.e. 3% NaTPB, and 10% OA) which produced a linear response with a slope of 19.9 ± 0.6 mV/decade from 1.0×10^{-6} to 1.0×10^{-2} mol L⁻¹ ($4.2 < \text{pH} < 8.0$). Pr³⁺, Dy³⁺, Nd³⁺, La³⁺, Tb³⁺, Cr³⁺, K⁺, Na⁺,

Mg^{2+} , Ca^{2+} , Pb^{2+} , Ni^{2+} , Cd^{2+} were evaluated as interfering ions and the maximum interference was observed from La^{3+} with a K_{MPM} of 8.6×10^{-4} .

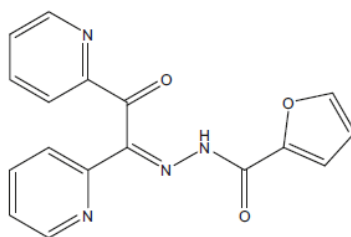


Figure 10. N'-(2-oxo-1,2-di(pyridin-2-yl) ethylidene)furan-2-carbohydrazide

The latest report on the development of a Gd^{3+} sensor is the work of Gadhari et al. [59], who used tetraazacyclododecane-1, 4,7,10-tetraacetic acid crown ether (Figure 11), as the first instance of a cyclic ion carrier for this purpose.

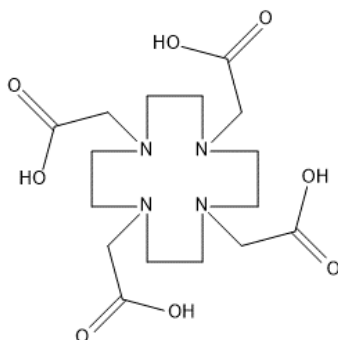


Figure 10. Tetraazacyclododecane-1,4,7,10-tetraacetic acid crown ether

They reported using multiwalled carbon nanotubes (MWCNT) to enhance the signal of the potentiometric PVC membrane sensor and recorded a slope of 20.05 ± 0.18 mV/decade from 1×10^{-8} to 1×10^{-2} mol L^{-1} and a limit of detection of $7.2 \pm 0.16 \times 10^{-9}$ mol L^{-1} .

3. CONCLUSION

Among the various selective Gd^{3+} ion sensors, almost all ionophores used have been acyclic ligands (i.e. (2-[(3-[(2-sulfanyphenyl)imino)-1-methylbutylidene]amino]phenyl hydrosulfide, Omeprazole, 6-methyl-4-[[1-(2-thienyl)methylidene]amino]3-thioxo-3,4-dihydro-1,2,4-triazin-5-(2H)-one, N-(2-Pyridyl)-N'-(4-nitrophenyl)thiourea, 2,6-bis-[1-{N-cyanopropyl,N-(2-methylpyridyl)}aminoethyl]pyridine,2-(4-phenyl-1,3-thiazol-2-yliminomethyl) phenol, 1-(4-nitrophenyl)-3-(4-phenylazophenyl)triazene) mostly with medium and soft donor atoms (N and S). There have also been some instances of acyclic ion carriers with medium and hard donor atoms, i.e. N and O (4-Methoxyphenyl cyanamide, N,N'-bis(methylsalicylidene)-2-aminobenzylamine, N'-(2-oxo-1,2-di(pyridin-2-yl)

ethylidene) furan-2-carbohydrazide). In all these cases the major mechanism for the selective complexation could be the formation of wrap-around complexes. The only exception to this trend is the application of tetraazacyclododecane-1,4,7,10-tetraacetic acid crown ether, which is a cyclic ligand with medium and hard donor atoms in which case the cavity size of the ionophore could be the further factor influencing its selectivity for the target ion.

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