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Review

A Review on Praseodymium Selective Electrochemical and Optical Sensors

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Abstract- Praseodymium is a rare earth element with various applications. The analysis of the levels of the compounds of this elements is very important and various instrumental techniques have been used for the determination of this element in various samples. Among the various techniques used to this end, electrochemical and optical sensors constitute a set of powerful tools for the determination of traces of Pr^{3+} ions in various samples and concentration ranges. The present review tends to provide an overview on the various potentiometric and optical sensors developed for this purpose, with a focus on the ionophores used and the composition of the sensing elements.

Keywords- Praseodymium; Ionophore; Ion selective electrode; Potentiometry; Sensor; Optode

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1. INTRODUCTION

Various modern applications (e.g. glass and ceramics, metallurgy, nuclear chemistry, and electronics) are dependent on the application of rare-earth elements, offering various unique properties [1-3].

As a rare-earth, praseodymium is a soft metal with anticorrosion characteristics, which is used in developing very resistant alloys applied in the construction of aircrafts engines. Pr is also used in glass and enamel formulations as a yellow coloring agent [4]. Pr/Ge alloys are present in superconductors [5].

Metallic praseodymium (Pr) is found in various forms. The soft, silvery, and ductile material has higher corrosion resistance in the presence of oxygen compared to Eu, La, Ce, and or Nd, yet a layer of oxide forms on its surface which easily chips away in the presence of air. This makes the maintenance of Pr targets difficult. Metallic Pr is stored in an atmosphere of inert gasses or in petroleum or in mineral oil. Given the nontoxicity of Pr, fabrication of praseodymium targets does not involve precautions. Praseodymium has a La-type double hexagonal structure with an ABAC stacking sequence of close-packed planes under ambient conditions [6,7].

The incremental demand has led to great need for the analysis of these species, which have very similar properties [1,2], and hence their analysis via conventional methods is very difficult [8,9]. Analysis of single lanthanides is commonly performed via physical approaches like emission spectrometry or a combination of chemical methods with physical techniques such as neutron-activation or X-ray fluorescence (XRF) techniques, yet total content of lanthanides present in rocks and minerals, is determined through gravimetric techniques. Analysis of traces of lanthanides remains difficult, given the interferences of different elements for one another or due to insensitivity issues. The application of isotope-dilution mass spectrometry (ID-MS), [10,11] neutron activation analysis [12] and HPLC-ICP-AES for the determination of lanthanides has been reported. Although ID-MS offers considerable sensitivity for the analysis of lanthanides, it suffers poor selectivity. Furthermore Pr, Tb, Ho and Tm cannot be analyzed via this technique since they do not have more than one multiple stable isotopes.

The chief problem in quantitative analysis for rare-earth ions is the selectivity of the analytical method. Various studies have been focused on the spectrophotometric analysis of these species using reagents such as Alizarin Red S, aluminon, Xylenol Orange, Arsenazo I, Arsenazo III, PAN and PAR, yet none has proven to be selective enough, to the extent that even the most selective methods require extractive pretreatment for removing interfering species.

A common technique used for the analysis of rare-earth ions in steel samples and minerals [2,13,14] is inductively-coupled plasma emission spectrometry (ICP-ES), yet the selectivity of the method was not always satisfactory, which led to the later development of inductively-coupled plasma emission derivative spectrometry (ICP-EDS) [15].

Voltametric analysis of lanthanides is very difficult due to the electrochemical behavior of these elements. The application Hg electrodes, produces poorly defined waves and requires very negative potentials [16]. In some research reducible organic dyes [17,18] were evaluated as complexing agents for lanthanide ions for use in voltametric analyses of these species. The results showed discrete reduction waves. Yet the determination of traces of lanthanides is not using sensitive electrochemical techniques like stripping voltammetry is still not very common, given the inherent difficulties [19,20].

Ion selective electrodes are devices with tunable properties and different advantages, the potentials of which are very well reflected by the application of these devices for various lanthanide ions (e.g. Ho^{3+}, La^{3+}, Yb^{3+}, Dy^{3+}, Tb^{3+}, Nd^{3+}, Lu^{3+}, and Sm^{3+}) [21–34]. This makes the application of potentiometric ion-selective [35-49] and optical [50-52] sensors a promising method for the analysis of praseodymium ions. The following lines tend to provide an overview on the development of various potentiometric and optical sensors for Pr^{3+} ions.

2. PRASEODYMIUM ELECTROCHEMICAL SENSORS

2.1. Potentiometric Pr ion Sensors

The first report on a PVC-based Pr^{3+}-selective electrode involved the application of N′-(pyridin-2-ylmethylene)benzohydrazide (Figure 1), was prepared using 30% wt. of PVC, 59% wt. of *o*-nitrophenyloctyl ether (NPOE), 7% wt. of the ionophore and 4% of sodium tetraphenyl borate. The membrane sensor had a linear response with a slope of 21.1 mV per decade of concentration in the concentration window of 1.0×10^{-6} to 1.0×10^{-2} M and a detection limit of around 115 ppb [35].

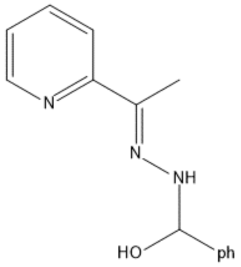

Figure 1. Chemical structure of N′-(pyridin-2-ylmethylene)benzohydrazide [35]

In another work a Pr^{3+} microsensor was developed using the same ionophore (i.e. N-(pyridin-2-ylmethylene) benzohydrazide). The developed microsensor was reported to have an identical membrane composition and its potential slope was similar to that of the membrane sensor, yet it had a wider linear response in the concentration range of 1.0×10^{-8} to 1.0×10^{-3} M and a detection limit of 1 ng/ml [36].

In another work a Pr^{3+} sensor was developed using PVC and polystyrene as binders with zirconium (IV) antimonotungstate. Membranes based on both binders worked well from 5×10^{-5} M to 1×10^{-1} M of Pr^{3+} ions and produced a near-Nernstian slope of 25.0 mV/decade of concentration, yet the polystyrene-based sensor had a faster response time, and the electrode was reported to possess a good selectivity profile [37].

In 2008 a novel Pr^{3+} sensor was prepared using N,N-bis(α-methylsalicylidene) diethylenetriamine (Figure 2) as an ionophore. The optimal membrane sensor contained 32% of PVC powder, 59% of NPOE, 6% of the ionophore and 3% of NaTPB and showed a response of 19.8±0.2 mV per decade from 1.0×10^{-6} to 1.0×10^{-2} M and a detection limit of 6.5×10^{-7} M [38].

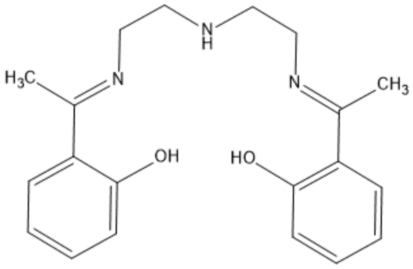

Figure 2. Chemical structure of N,N-bis(α-methylsalicylidene)diethylenetriamine [38]

Another report on a Pr^{3+} sensor introduced a PVC membrane electrode including N′1,N′-2-bis(2-oxo-1,2-diphenylethylidene) ethanedihydrazide (Figure 3) with a 19.6±0.4 mV per decade slope from 1.0×10^{-6} to 1.0×10^{-2} M and a limit of detection (LOD) as low as 4.3×10^{-7} M [39]. The optimal electrode composition was reported to be 30% of PVC powder, 66% of acetophenone (AP), and 2% of the ionophore and NaTPB each. The matched potential selectivity coefficients were determined to be Yb^{3+}:3.2×10^{-4}, Fe^{3+}:1.0×10^{-3}, Tm^{3+}:1.0×10^{-3}, Eu^{3+}:2.7×10^{-3}, La^{3+}:8.6×10^{-3}, Er^{3+}: 4.8×10^{-3}, Ce^{3+}: 2.3×10^{-3}, Dy^{3+}: 7.4×10^{-4}, Tb^{3+}: 4.2×10^{-4}, Cr^{3+}: 8.2×10^{-3}, Sm^{3+}: 4.3×10^{-4}, Ca^{2+}: 5.8×10^{-3}, Nd^{3+}: 3.7×10^{-4}, Co^{2+}: 1.6×10^{-3}, Lu^{3+}: 4.3×10^{-4}, Ni^{2+}: 6.7×10^{-4}, Ho^{3+}: 6.4×10^{-4}, Pb^{2+}: 7.6×10^{-4}, Gd^{3+}: 7.2×10^{-3}, Na^{+}: 2.4×10^{-4}. Based on the data the maximum interfering effects were caused by La^{3+}, Gd^{3+} and Cr^{3+}.

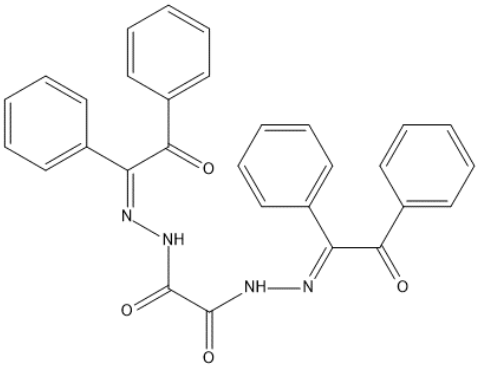

Figure 3. Chemical structure of N'_1,N'_2-bis(2-oxo-1,2-diphenylethylidene) ethanedihydrazide [39]

In 2009, Ganjali et al reported developing a carbon paste electrode for Pr^{3+} ions [40] using multiwalled carbon nanotubes, 1-n-butyl-3-methylimidazolium tetrafluoroborate, and N’-

(pyridin-2-ylmethylene) benzohydrazide (Figure 1) which was used in construction of Pr^{3+} sensors before [35,36]. The presence of the ionic liquid was reported to improve the response of the developed CP and the optimal composition was mentioned to be 30% of the ionic liquid ([bmim]BF_4), 17% of the ion carrier, 38% of graphite powder and 15% of multiwalled CNT (MWCNT), which led to a Nernstian response of 19.9±0.3 mV/decade of concentration in the concentration window of 2.5×10^{-7} and 1.0×10^{-1} M. Except for Pb^{2+} ions which produced an selectivity coefficient of 1.0×10^{-2} for the rest of the tested ions the selectivity coefficients were lower than 2.1×10^{-3} (Gd^{3+} and Yb^{3+}).

In another research two PVC-membrane electrodes were prepared suing 1,3-diphenylpropane-1,3-diylidenebis(azan-1-ylidene)diphenol (L1) and N,N′-bis(pyridoxylideneiminato) ethylene (L2). The former ionophore was reported to have proper response and the best composition, based on L1, was reported to be 32.4% of PVC, 64.8% of o-NPOE: 1.7% of the ionophore and 1.1% of NaTPB. This membrane had a linear response with a Nernstian slope of 20.0±0.3 mV $decade^{-1}$ from 1.0×10^{-8} to 1.0×10^{-2} M (detection limit: 5.0×10^{-9} M) [41].

Faridbod et al [42] used developed another modified CPE for the analysis of Pr^{3+} ions using MWCNT, an ionic liquid (IL) and an ionophore which had already been used in developing a liquid membrane sensor for the same analyte (i.e. N,N-bis(a-methylsalicylidene) diethylenetriamine (Figure 2) [38]). The optimal composition of the carbon paste was reported to be 25% wt. of the ionophore, 25% of the ionic liquid, 45% of graphite powder, and 5% of MWCNT, which lead to e Nernstian response of 20.1±0.4 mV per decade from 1.0×10^{-6}-1.0×10^{-2} M. The selectivity coefficients of the electrode for various ions were reported as (Na^{+}:1.3×10^{-4}, Gd^{3+}:4.2×10^{-3}, K^{+}:2.0×10^{-4}, Yb^{3+}: 6.6×10^{-3}, Nd^{3+}: 3.7×10^{-3}, Tb^{3+}: 5.7×10^{-3}, Ho^{3+}: 8.6×10^{-4}, La^{3+}: 1.3×10^{-3}, Ca^{2+}: 7.4×10^{-4}, Sm^{3+}: 5.0×10^{-3}, Cu^{2+}: 2.2×10^{-4}: Dy^{3+}: 4.1×10^{-3}, Pb^{2+}: 2.5×10^{-4}, Lu^{3+}: 7.0×10^{-4}, Fe^{3+}:1.5×10^{-4}, Eu^{3+}: 2.0×10^{-3}, Zn^{2+}: 4.2×10^{-4}, Ce^{3+}: 4.6×10^{-3}, Tm^{3+}: 6.4×10^{-3}: Er^{3+} 5.5×10^{-4}), indicating the considerable selectivity of the developed sensor for different common interfering ion.

In 2011, a praseodymium ion sensor was reported based on N-N'-o-phenylene-bis(salicylideneimine) (Figure 4) and two different binders, i.e. epoxy resin and PVC. It was reported that a composition of 50% of the ionophore and 5% of the binding material (epoxy resin) showed the best response behavior. Using the epoxy resin binder, the slope of the electrode response was 19.9 mV/decade from 1.0×10^{-6} to 1.0×10^{-1} M [43]. Using PVC, the sensor had less sensitivity (a slope of 14.0 mV/decade), while the response was linear in the same range of 1.0×10^{-6} to 1.0×10^{-1} M. The electrodes were also reported to have good selectivity profiles against alkaline, alkaline earth and transition metal ions.

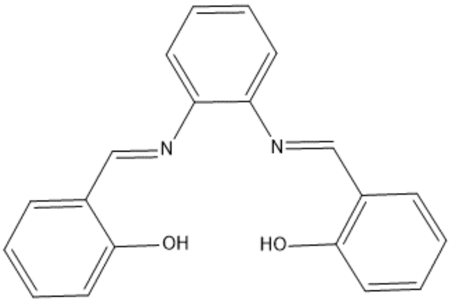

Figure 4. Chemical structure of N-N'o-phenylene-bis(salicylideneimine) [43]

In the same year Zamani et al [44] reported a PVC-based Pr^{3+} membrane using N,N′-bis(4-hydroxysalicylidene)-1-3- phenylenediamine (Figure 5) as the ionophore, together with 2% wt. of NaTPB, 65% wt. of BA and 30% wt. of PVC powder. The slope of the calibration curve of the sensor was 19.8±0.4 mV per decade of concentration from 1.0×10^{-6} to 1.0×10^{-2} M and a detection limit of 5.7×10^{-7} M. The selectivity coefficients of the sensors for various interfering ions were reported to be Pr^{3+}: 5.6×10^{-6}, La^{3+}: 2.0×10^{-4}, Sm^{3+}:1.1×10^{-4}, Yb^{3+}: 9.6×10^{-3}, Ho^{3+}: 7.3×10^{-3}, Nd^{3+}:4.2×10^{-3}, Eu^{3+}: 4.1×10^{-3}, Lu^{3+}:3.5×10^{-3}, Tm^{3+}: 3.3×10^{-3}, Er^{3+}: 1.4×10^{-3}, Gd^{3+}:9.1×10^{-2}, Tb^{3+}:8.7×10^{-2}, Dy^{3+}:8.3×10^{-2}, Co^{2+}: 6.6×10^{-2}, Ni^{2+}: 5.4×10^{-2}, $Fe^{3+}<10^{-2}$, $Cr^{3+}<10^{-2}$, $Na^{+}<10^{-2}$, $K^{+}<10^{-2}$, $Ca^{2+}<10^{-2}$, $Pb^{2+}<10^{-2}$, indicating the applicability of the sensor in the presence of different commonly occurring species.

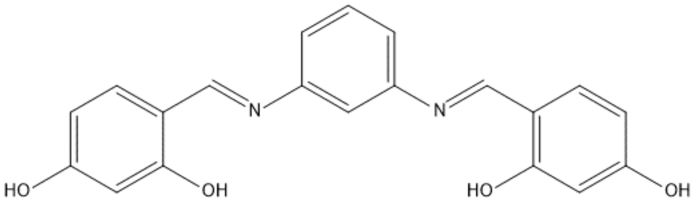

Figure 5. Chemical structure of N, N′-bis(4-hydroxysalicylidene)-1-3- phenylenediamine [44]

One year later Pourjavid et al [45] used a furan-triazole derivative (i.e. 3-nitro-4-amino-5-naphtho[2,1-b]furan-2-yl-4H-1,2,4-triazole-3-thiol (Figure 6)) to develop a Pr^{3+}-selective sensor, with a Nernstian slope of 20.5 ± 0.2 mV/decade of concentration from 1.0×10^{-7} to 5.0×10^{-1} M. The optimal polymeric membrane composition included 30% wt. of PVC powder, 59% wt. of *o*-NPOE, 8%wt of the ion carrier, and 3%wt. of NaTPB. The highest interference was caused by $Nd^{3+}$and Ce^{3+} with respective selectivity coefficients of 6.3×10^{-3} and 6.1×10^{-3} [45].

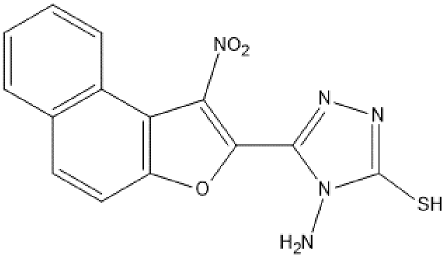

Figure 6. Chemical structure of 3-nitro-4-amino-5-naphtho[2,1-b] furan-2-yl-4H-1,2,4-triazole-3-thiol [45]

Another ionophore used for the construction of a Pr^{3+}ion selective sensor was 2,3,4,5-tetra-(4-pyridiyl)-thiophene (TPT) [46]. A membrane containing 3% of the ionophore, 3%wt. of NaTPB, 64%wt. of nitrobenzene together with 30% of PVC powder had a linear response with a slope of 20.2±0.5 mV per decade of concentration in the concentration range of 1.0×10^{-6} to 1.0×10^{-2} M (Detection limit: 5.3×10^{-7} M) [46].

In another report a modified nanocomposite composed of MWCNT/nanosilica and bis(salicylaldehyde)thiocarbohydrazone (Figure 7) was used to prepare a Pr^{3+}electrode. The optimal carbon paste composition contained 3% of bis(salicylaldehyde)thiocarbohydrazone, 30% wt. of paraffin oil, 2% of MWCNT, 0.3% of nanosilica, and 64.7% of graphite powder. The slope of the electrode response was 19.6±0.2 mV decade of concentration from 1.0×10^{-8} to 1.0×10^{-2} M and a detection limit of 8.5×10^{-9} M [47].

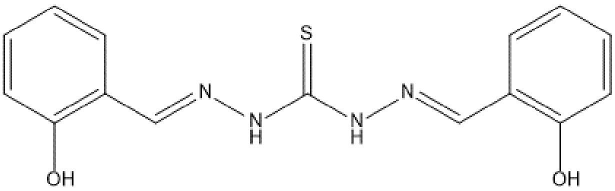

Figure 7. Chemical structure of bis(salicylaldehyde) thiocarbohydrazone [47]

The same ionophore was used to construct a PVC-based membrane sensor for Pr^{3+}. The optimal composition included 3% wt. of the ionophore, 64% wt. of NB, 3% wt. of NaTPB and 30% wt. of PVC powder and had a linear response with a slope of 19.5±0.7 mV per decade from 1.0×10^{-6} M to 1.0×10^{-2} M [48].

Rather recently N, N-bis(a-methylsalicylidene) diethylenetriamine (Figure 2) was used to develop a Pr^{3+} sensor with a slope of 19.6±0.1 mV per decade from 1.0×10^{-7} to 1.0×10^{-2}. The optimal electrode composition was described as containing 10% wt. of N and S containing porous graphite (NSPG, 15% wt. of the ionophore, 30% of an ionic liquid and 45% graphite powder [49].

2. PRASEODYMIUM OPTICAL SENSORS

Optical sensors like electrochemical sensors are also useful tools in determination of ionic species [50,51]. Some optical sensors have been introduced for Pr^{3+} ions in the literature [52-54]. The first instance is the work of Ganjali et al in 2013 who used (E)-2-(1-(4-hydroxy-2-oxo-2H-chromen-3-yl) ethylidene) hydrazinecarbothioamide (Figure 8) to construct a sensor for the determination of Pr^{3+} in water-acetonitrile solutions via fluorescence spectroscopy. They reported that the 1:1 binding of E)-2-(1-(4-hydroxy-2-oxo-2H-chromen-3-yl) ethylidene) hydrazinecarbothioamide with Pr^{3+} in a (9/1:v/v) mixture of acetonitrile and water leads to enhancement in its fluorescent properties which was used for the determination of Pr^{3+} from 1.6×10^{-7} to 1.0×10^{-5} M (LOD: 8.3×10^{-8} M) with good selectivity [52].

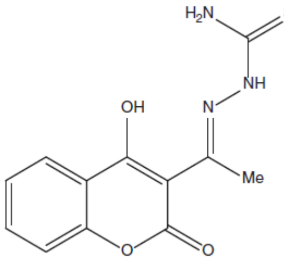

Figure 8. Chemical Structure of (E)-2-(1-(4-hydroxy-2-oxo-2H-chromen-3-yl)ethylidene) hydrazinecarbothioamide [52]

In another research an optical chemical sensor (optode) was designed through impregnating N,Ń-bis (salicylidene)-1, 3-ethylenediamine (Figure 9) into a paste of mesoporous SBA-15. The optical sensor was used for the determination of Pr^{3+} ion via UV/Vis spectrophotometry. To fabricate the sensor the indicator was dip-coated with a triacetylcellulose. The resulting optode had a linear response from 10 to 190 ng mL^{-1} and its detection limit reached as low as 5.0 ng mL^{-1} and it had good selectivity for the analyte in the presence of cerium, niobium, lanthanum, chromium, zinc, mercury (II) and iron (II) ions [53].

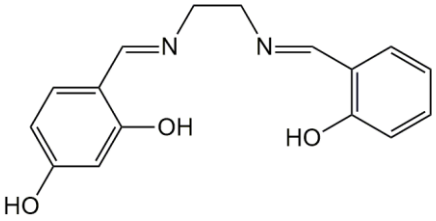

Figure 9. Chemical Structure of N, Ń-bis (salicylidene)-1, 3-ethylenediamine [53]

The latest report on an optical sensor for Pr^{3+} ions using a luminous molecular receptor C4DS based on calix[4]arene conjugate bearing dansyl fluorophore with hydrazine carbonyl linkage (Figure 10). The sensor was reported to be also applicable for the determination f iodide ions. The lower detection limits of the respective chemosensor for Pr^{3+} and I^-were 3.571 and 2.439 nM, and its response to the two ions was linear in the two concentration windows of 0–135 nM and 0–120 nM [54].

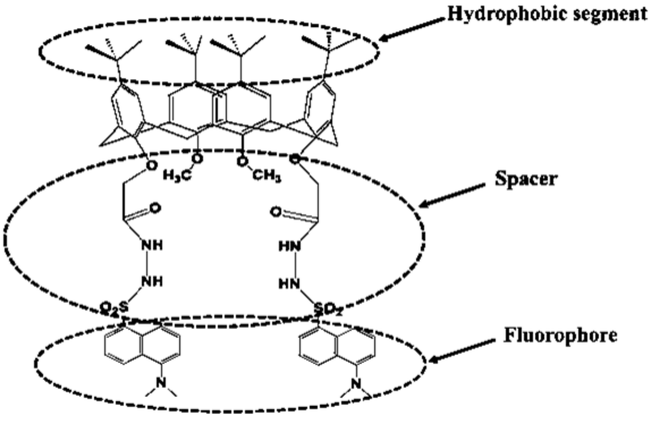

Figure 10. Chemical Structure of the fluorophore [54]

4. CONCLUSION

Among the various polymeric membrane and carbon paste electrodes used all have been baed on acyclic ligands, (i.e. N′-(pyridin-2-ylmethylene) benzohydrazide, N,N-bis(α-methylsalicylidene) diethylenetriamine, N′1,N′2-bis (2-oxo-1,2-diphenylethylidene) ethane dihydrazide, 1,3-diphenylpropane-1,3-diylidenebis(azan-1-ylidene)diphenol, N-N'-o-phenylene-bis(salicylideneimine), N,N′-bis(4-hydroxysalicylidene)-1-3- phenylenediamine, 3-nitro-4-amino-5-naphtho[2,1-b] furan-2-yl-4H-1,2,4-triazole-3-thiol, bis(salicylaldehyde) thiocarbohydrazone) a pattern that is also observed in the case of optical electrodes developed for Pr^{3+} ions (using (E)-2-(1-(4-hydroxy-2-oxo-2H-chromen-3-yl)ethylidene) hydrazine carbothioamide and N,Ń-bis (salicylidene)-1, 3-ethylenediamine). This indicates that the formation of wrap around complexes can be considered as the main mechanism for the formation of complexes between the analyte and these ion carriers. The phenomena can be regarded as the major reason behind the selectivity profiles of the sensors based on these ionophores.

Acknowledgements

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