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Potentiometric determination of polyether ionophores growth promotors in animal feed preparations

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Abstract

The construction and electrochemical response characteristics of four polyvinylchloride (PVC) membrane sensors for the determination of monensin (MN) and salinomycin (SL) were described. The membranes were prepared using 1 wt.% drug, 44 wt.% nitrophenyl octyl ether, 53 wt.% PVC and 2 wt.% lipophilic additive which is the anionic potassium tetra (4-chlorophenyl) borate in sensors 1 and 2 and the cationic nicklo-phenanthroline in sensors 3 and 4. Sensor 1 and sensor 2 show linear responses over concentration range of 10^{-3}–10^{-5} M drug with cationic slopes of 52.3 and 54.1 mV per concentration decade, respectively. On the other hand sensor 3 and sensor 4 show linear responses over concentration range 10^{-4}–10^{-5} M drug with anionic slopes of 28.1 and 29.7 mV per concentration decade, respectively. The 4 sensors were successfully applied to the determination of MN and SL in their pharmaceutical products (Premix) with average recoveries of 98.4–100.4 $\pm$ 2.41–1.97% for sensors 1 and 2 and 97.6–98.8 $\pm$ 3.16–3.07% for sensors 3 and 4. The obtained results were compared reasonably well with the data obtained using the USP method (2000). The proposed sensors were also applied for the direct determination of both drugs in animal feed preparations without prior treatment in low levels; 10–1000 μg per 5 g feed. The sensors were successfully used to follow up the drugs concentration in the presence of other growth promotors, other antibiotics and the coexisting fatty acids.

Keywords: Polyether ionophores potentiometry; Tetrachlorophenylborate; Nicklo-phenanthroline; Animal feed preparations

1. Introduction

Monensin (MN) and salinomycin (SL) belong to the polyether antibiotics, a group of naturally appearing ionophore substances that are produced by certain strains of streptomyces species [1,2].They also act as growth promotors via increasing feed conversion efficiency [3]. Their main therapeutic application is in veterinary medicine as food additives; added to animal feed at a concentration of 10–120 mg kg^{-1} and to poultry feed in a concentration of 80 ppm [4].

Microbiological assays have been adopted as their official analytical methods in Japan [5] and other countries [6,7]. A variety of chemical analyt-

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ical methods have been reported for the determination of MN and SL. Colorimetric procedure using vanillin as a chromogenic reagent [8,9], spectrodensitometric [10,11] and flow injection analysis [12] have been recommended. These methods are not suitable for the analysis of feed preparations due to their high blanks values. Different liquid chromatographic methods have also been recommended [13–15]. One HPLC procedure has been reported for the separation of MN and SL, using electrospray mass-spectrometry [16].

MN has also been determined by a voltametric method between two immiscible electrolyte solutions [17]. Many authors [18–22] suggested potentiometric membrane sensors based on polyether ionophores and their derivatives for the determination of some inorganic ions, surfactants and other purposes.

In the present work, both the cationic and anionic properties of MN and SL have been fully studied. MN and SL were found to react well with anionic and cationic exchangers with the formation of highly lipophilic and remarkably stable ion association complexes, which are suggested to be used as electroactive materials in polyvinylchloride (PVC) matrix membrane sensors. The investigated sensors determine MN and SL in premix and feed-preparations directly, simply, rapidly and selectively.

2. Experimental

2.1. Materials and reagents

Deionized water was used throughout the procedure and all chemicals used were of analytical reagent grade. Pure MN and SL were provided by Sigma Co. The lipophilic additives k-Tp ClPB and phenanthroline were obtained from Fluka. Ni–Ph was prepared by dissolving 100 mg of phenanthroline in 20 ml of 10^{-2} M nickel (II) chloride, followed by 5 drops of ethanol to keep the solution clear. PVC powder, 2-nitrophenyloctylether (NPOE) and dibutyl sebathate (DBS) were purchased from Aldrich. Standard solutions were freshly prepared with deionized water; MN and SL are stable in solution for up to 4 weeks. Pharmaceutical preparations (premixes) were obtained from the local market, containing 10% of MN or SL; their guaranteed analysis was given in Table 1. In addition, poultry solid feed samples were used.

2.2. Apparatus

Measurements were made at 25 ± 1 °C with an Orion Ionanalyser (Model 920 digital pH/mV meter) using drug-PVC membrane sensor in conjunction with an Orion single junction Ag–AgCl reference electrode (Model 90-02). An Orion combination pH electrode (Model 91-02) was used for pH adjustment.

2.3. Monensin and Salinomycin–PVC membrane sensors

The polymeric membranes were prepared by mixing 5.6 mg MN or SL, 296.8 mg PVC, 246.4 mg NPOE and 11.2 mg lipophilic additive with 3 ml THF. The resulting homogenous syrup was poured into a 50 mm diameter ground glass casting ring and the solvent was allowed to evaporate

Table 1
Composition of premixes[a] containing 10% MN or SL

Premix	Constituents[b]
Ca^{2+}	4.7–5.7%
P^{3+}	4.0%
Salt	16.6–19.9%
K^{+}	1.5%
Mg^{2+}	0.2%
I_2	48 ppm
Cu^{2+}	25 ppm
Ce^{2+}	13.3 ppm
Zn^{2+}	540 ppm
Vitamin A	100 000 IU/lb
Vitamin D_3	25 000 IU/lb
Vitamin E	25 IU/lb

[a] Obtained from Lilly Co., Cairo, Egypt, by personal communication.

[b] This is the guaranteed analysis. In addition to insert approved diluents include, cane molasses, beef molasses, soya bean, corn cope fraction, calcium phosphate, animal fats, ZnO, $FeCO_3$, $CuSO_4$, $CaCO_3$, sodium selenate, ethylenediamine dihydroiodide vitamin A, E and D_3 supplements.

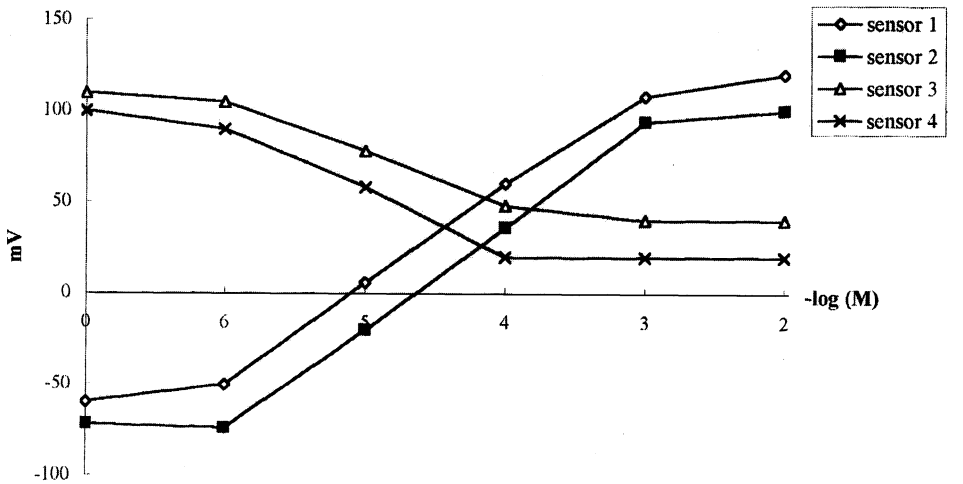

Fig. 1. Profile of the potential in mV to the $-\log$ concentration using sensors 1, 2, 3 and 4.

off slowly at room temperature over a period of 48 h. A semi-transparent flexible membrane with a thickness of 0.25 mm was obtained. A disc (8 mm diameter) was cut using a cork borer and pasting using THF, to an interchangeable PVC tip which was clipped onto the end of the electrode glass body [23,24]. Solutions of 1:1 KCl and drug 1×10^{-2} M each was used as internal reference solution and Ag–AgCl wire (1 mm diameter) was used as internal reference electrode. The sensor was preconditioned after preparation by soaking it for 10–24 h in 10^{-2} M drug solution and stored in 10^{-2} M KCl solution when it is not in use. The sensor was washed with deionized water and covered with tissue-paper between measurements.

Sensor calibration was carried out by measuring the potential of 10^{-6}–10^{-2} M drug solutions, starting from the low to the high concentrations. The potentials were plotted as a function of minus logarithm drug concentrations; Fig. 1. The following regression equations were used to calculate unknown concentrations of the analytes:

$$E_1 = 52.3(-\log[\mathrm{MN}]) + 268.6$$

$$E_2 = 54.1(-\log[\mathrm{SL}]) + 261.0$$

$$E_3 = 28.1(-\log[\mathrm{MN}]) + 65.5$$

$$E_4 = 29.7(-\log[\mathrm{SL}]) + 119.0$$

where E is the measured potential in mV.

Sensor life span was examined by repeated monitoring of the slope of drug calibration curve periodically.

Repeatability was measured by immersing the sensor alternatively into 10^{-5} and 10^{-4} M drug solutions at 25 °C. In addition, the reproducibility of the proposed procedures was examined using 10^{-5}–10^{-3} M for sensors 1 and 2 and using 10^{-5}–10^{-4}.

Selectivity coefficients of each sensor were determined in the presence of nine interfering substances (Table 3). The separate solution method [25] was used and calculated from the rearranged Niclosky equation:

$$\mathrm{Log}\, K^{\mathrm{pot}}_{\mathrm{drug,M}} = [E_{\mathrm{drug}} - E_{\mathrm{M}}/S] + [1 + Z_{\mathrm{drug}}/Z_{\mathrm{M}}]\log[\mathrm{drug}]$$

where: E_{drug} is the potential measured in 10^{-4} M drug solution, E_{M} is the potential measured in a 10^{-4} M interferent solution, Z_{drug} and Z_{M} are the charges of the drug and interfering ion, respectively. S is the slope of the electrode calibration plot.

2.4. Determination of the drugs in pharmaceutical formulations

The contents of three Premix packets (their

composition is given in Table 1) were mixed and a quantity of the mixed contents corresponding to about 0.069 and 0.077 g of MN and SL, respectively, were transferred to a 100-ml beaker. About 80 ml of water was added and the beaker was placed on the shaker for 1 h. The contents of the beaker were accurately transferred to a 100-ml volumetric flask and completed to the mark with water; preparing 10^{-3} M aq. Drug solution. One millilitre aliquot of this solution was diluted to 100 ml and PVC membrane sensor in conjunction with an Ag–AgCl reference electrode was immersed in the solution. The potential was measured before and after the addition of 0.1 ml of 10^{-2} M drug solution. The original concentration of the drug in the test solution was measured using the standard addition equation [26].

2.5. Application to animal feed preparations

Standard experimental feed for chicken was used; its raw materials were corn, rice, bran and soya bean. A large quantity of fatty acids were also contained. Five grams of food was used with the addition of standard drug powder in the range of 10–1000 μg. It was diluted with water, mixed and the analysis was completed as in Section 2.4.

3. Results and discussion

The increasing importance of feed additives in animal resources makes the manufacture of ion-selective electrodes for their determination, an area of interest. The advantages they offer over other analytical procedures are: low cost, simplicity, suitability for testing samples in animal feed and in Premix directly, without prior treatment.

3.1. Monensin and Salinomycin ionophores

Both MN and SL are acidic ionophores carrying carboxylic groups [27]. They are recommended to be polyether ionophores that act as neutral carriers not charged one [28]. They have low water-solubility and this can be explained by its cyclic structure and the hydrophobic function being located on the exterior surface [29]. The partially dissociated molecule of either drug is able to form negatively and positively charged complexes in the membrane phase [28]. This mixed mode was studied here in detail and used for the investigation of highly selective electrodes with both anionic and cationic additives.

$$\mathrm{SL{-}H} \leftrightarrows \mathrm{SL^-} + \mathrm{H^+}$$

$$\mathrm{MN{-}H} \leftrightarrows \mathrm{MN^-} + \mathrm{H^+}$$

where $\mathrm{MN^-}$ (or $\mathrm{SL^-}$) is the charged form and MN–H (or SL–H) is the neutral form. Both forms may build complexes with ions in the membrane phase leading to a negatively charged $(\text{anion}{-}\mathrm{MNH})^-$ and a positively-charged $(\text{cation}_2{-}\mathrm{MN})^{2+}$

3.2. Membrane characteristics and preliminary studies of complexation

Plasticized PVC membranes with anionic and cationic lipophilic additives were prepared and electrochemically evaluated as prospective sensors for MN and SL drugs according to IUPAC standards [25]. Table 2 shows that the four proposed sensors consisted of 1% drug, 44% plasticizer, 53% PVC and 2% lipophilic additive, displaying ideal performance characteristics.

Typical calibration graphs for the proposed sensors are shown in Fig. 1. Reasonably stable behaviour of the sensors was maintained during at least 6 weeks.

At 25 °C, the sensors display linear responses for MN and SL concentrations over the range 10^{-3}–10^{-5} and 10^{-4}–10^{-5} M, using anionic and cationic additives, respectively; Fig. 1. They show calibration slopes of 52.3, 54.1, 28.1 and 29.7 ($n > 6$) for the sensors MN–K–TpClPB, SL–K–TpClPB, MN–Ni–Ph and SL–Ni–Ph, respectively (Table 2). Repeated calibrations of the sensors over a period of 6 weeks with drug solutions show potential and slope stabilities within ± 1 mV decade^{-1}.

The accuracy of the proposed procedures ranged from 99.7–100.8% (Table 2). Repeatability was assured by immersing each sensor alternatively into 10^{-5} and 10^{-4} M solutions, where the

RSD ranged between 2.15 and 2.84%. The reproducibility was evaluated over a period of 3 weeks, during which the RSD values were 2.9–3.5%

The chemical composition and stoichiometry of the drug-additive complexes were identified by elemental analysis and IR spectra. Complexes of 1:1 or 1:2 ratios were found between each drug and anionic or cationic lipophilic additive, respectively.

3.3. Effect of lipophilic additives

It has been recommended that the addition of ionic sites for neutral carrier-based ion-selective electrodes is beneficial. It reduces the interference by lipophilic counter ions and creates permselectivily [30].

In the present work, it was found that the anionic additive led to the best performance. It reduced the response time, lowered the electrical membrane resistance and gave rise to improved selectivity (Table 2).

3.4. Effect of PVC and plasticizer

Similar results were obtained with PVC or PVC–COOH, where only a small portion of the carboxyl groups is ionised. It is noteworthy to mention that the life span of the membrane containing PVC–COOH is much longer than those containing PVC (10 weeks in comparison to 6 weeks in the case of PVC).

The potentiometric response of the investigated sensors was not influenced by the polarity of the membrane medium, which was in turn defined by the dielectric constants of the major membrane components. PVC matrix membrane incorporating drug with two different plasticizers having dielectric constants over the range of 4–24; namely DBS and NPOE were prepared, tested and the results were more or less the same. These data are in good agreement with previous reports showing that the response characteristics and selectivities of ionophores with carboxyl groups did not depend on the negatively charged ($-COO^-$) group or the polarity of the medium [28,31].

Table 2
Potentiometric response characteristics of the investigated membrane sensors

Parameter	Value[a]			
	Sensor 1	Sensor 2	Sensor 3	Sensor 4
	MN–K–TpClPB	SL–K–TpClPB	MN–Ni–Ph	SL–Ni–Ph
Concentration range (M)	10^{-3}–10^{-5}	10^{-3}–10^{-5}	10^{-4}–10^{-5}	10^{-4}–10^{-5}
Slope, (mV decade^{-1}) ± RSD	52.3 ± 0.3	54.1 ± 0.2	28.1 ± 0.4	29.7 ± 0.2
Correlation coefficient '*r*'	0.998	0.998	0.997	0.997
Intercept (mV) ± RSD	268.6 ± 0.9	261.0 ± 0.8	65.5 ± 0.4	119.0 ± 0.5
LOD (M)	10^{-6}	10^{-6}	10^{-6}	10^{-6}
LOQ (M)	10^{-5}	10^{-5}	10^{-5}	10^{-5}
Working range (pH)	4–10	4–10	4–10	4–10
Response time (s)	10	10	30	30
Accuracy (%)	100.1	100.8	99.9	99.7
Precision (RSD)				
Repeatability	2.36	2.15	2.84	2.71
Reproducibility[b]	3.1	2.9	3.5	3.2

[a] Mean of five measurements.
[b] Over a period of 3 weeks.

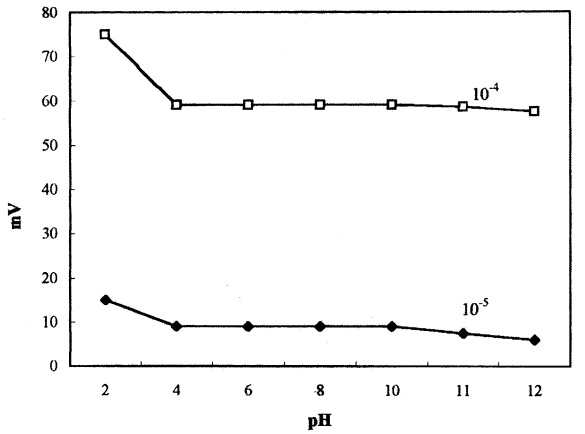

Fig. 2. Effect of pH on the response of sensor 1.

3.5. Soaking and response time

Freshly prepared electrodes must be soaked in 10^{-2} M drug solution to activate the surface of the membrane; depending on the diffusion and equilibration at the interface. The presoak times were overnight. The response times were nearly instantaneous in relatively concentrated solutions, while in dilute solutions, about 20 s were necessary to reach stable potential reading.

Nevertheless, continuous soaking of the electrodes in 10^{-2} M drug solution, affects negatively their response to the analyte. This may be attributed to leaching of the ionophore.

The response time (t_{95}) of the proposed sensors was tested by measuring the time required to achieve a 95% steady potential for 10^{-4} M and 10^{-5} drug solutions when their concentrations were rapidly increased by one decade. Fairly short response times of 10 and 30 s were obtained for sensors with anionic and cationic additives, respectively (Table 2).

3.6. Effect of pH

The pH dependence of the investigated sensors was examined using 10^{-4} M and 10^{-5} M drug solutions. Stable response over the pH range 4–10 was obtained (Fig. 2). Adjustment of pH was performed using dilute LiOH and HCl. Below pH 4, the sensor response increases with the increase of the analyte acidity; at such high acidity, the membrane may extract H^+ ions.

3.7. Selectivity of the electrodes

The potentiometric selectivity coefficient ($K^{pot}_{drug,interferent}$) of the proposed sensors were evaluated using the separate solution method [25] with 10^{-4} M concentration level of drug and interferents, which may be expected to be present in animal feed preparations. These interferents include lasalocid, flumequin, enrofloxacin, terramycin, penicillin sodium stearic acid, oleic acid

Table 3
Selectivity coefficient ($K^{pot}_{drug,interferent}$) of the four proposed PVC sensors

Interferent	$K^{pot}_{drug,interferent}$			
	Sensor 1	Sensor 2	Sensor 3	Sensor 4
	MN–K–TpClPB	SL–K–TpClPB	MN–Ni–Ph	SL–Ni–Ph
Monensin	–	6.3×10^{-2}	–	5.8×10^{-1}
Salinomycin	7.2×10^{-2}	–	7.1×10^{-1}	–
Lasalocid	3.1×10^{-2}	3.9×10^{-2}	8.3×10^{-2}	1.1×10^{-1}
Flumekin sod	1.1×10^{-4}	1.6×10^{-4}	4.2×10^{-3}	4.1×10^{-3}
Enrofloxacin sod	1.5×10^{-4}	1.1×10^{-4}	6.3×10^{-3}	3.7×10^{-3}
Terramycin sod	1.3×10^{-4}	1.7×10^{-4}	2.4×10^{-3}	1.4×10^{-3}
Penicillin sod	1.8×10^{-4}	1.7×10^{-4}	1.9×10^{-3}	5.1×10^{-3}
Stearic acid	1.3×10^{-4}	2.1×10^{-4}	8.9×10^{-4}	6.4×10^{-4}
Oleic acid	1.8×10^{-4}	2.7×10^{-4}	8.8×10^{-4}	7.6×10^{-4}
Palmetic acid methyl ester	1.7×10^{-4}	2.7×10^{-4}	9.7×10^{-4}	9.1×10^{-4}

Table 4
Determination of MN and SL in their pharmaceutical products using the proposed PVC matrix membrane sensors

Pharmaceutical product	Nominal drug content	Recovery ± C.V.%				
		Sensor 1[a]	Sensor 2[a]	Sensor 3[a]	Sensor 4[a]	Official method[b]
Rumensin premix, Lilly Co.	10% Monensin, Sodium	98.4 ± 2.41	99.9 ± 2.18	97.6 ± 3.16	98.8 ± 3.07	100.2 ± 0.78
B.N.064147		$F = 9.24$	$F = 7.80$	$F = 15.62$	$F = 15.13$	–
Sacox premix, Aventis Co.	10% Salinomycin, Sodium	100.4 ± 1.97	99.5 ± 2.31	98.7 ± 3.19	98.7 ± 3.13	99.8 ± 0.71
B.N.09314		$F = 7.76$	$F = 10.48$	$F = 19.68$	$F = 18.94$	–

Tabulated F value is 19.3 at 95% probability.
[a] Average of five measurements.
[b] Average of three measurements.

Table 5
Drug recoveries in synthetic mixtures with standard experimental feed

Added drug to the feed (μg)	Drug recovery (%)[a]			
	Sensor 1	Sensor 2	Sensor 3	Sensor 4
10	101.6	100.7	99.7	99.8
100	100.9	100.1	98.9	100.2
1000	99.1	99.8	100.3	101.4

Standard experimental feed, always equals 5 g.
[a] Mean of five determinations; maximum deviation is 2.36.

and palmetic acid methylester, (Table 3) The results obtained (Table 3) show that the selectivity of the sensors is in the order:

$$\text{Drug–K–T}_\text{p}\text{ClPB} > \text{drug–Ni–Ph}$$

The strong electrostatic interactions between the negative ionic charge and the dipoles of the studied ionophores, MN and SL, are primarily responsible for selectivity improvement, with the anionic additive, K–TpClPB.

The sensors with anionic additives (1 and 2) show high selectivity towards 10 interferents, while those with cationic additives (3 and 4) show reasonable selectivity; Table 3. Accordingly, the proposed sensors can be successfully used to follow up the drug concentration in the presence of other growth-promotors, other antibiotics and co-existing fatty acids.

3.8. Analytical applications

Utility, sensitivity and selectivity of the proposed sensors were verified by the direct determination of MN and SL in Premix (Table 4) and by determining 10–1000 μg of either drug in synthetic mixtures with 5 gm of standard experimental food using the standard addition technique (Table 5). Results with average recoveries of 98.4–100.4 ± 2.41–1.97% were obtained for Premix analysis. These results were compared with the data obtained using the HPLC pharmacopoeial method [32]; Table 4. The F-test revealed no significant difference between the means and variances of the two sets of results. The good agreement between potentiometric and chromatographic data demonstrates the applicability of the proposed sensors for routine analysis of real sam-

ples containing MN and SL without prior separation.

4. Conclusion

The official microbiological methods [5–7] are complicated, time consuming and non specific. Also the colorimetric [8,9], spectrofluorimetric [10,11] and flow injection [12] analysis are not suitable for the analysis of feed preparations due to their high blanks values. In addition, the liquid chromatographic methods used pre or post column derivatization due to the possibility of direct drug detection by UV, fluorescence or electrochemical detectors.

However, the proposed potentiometric procedures are sensitive, offer fast response, low cost and highly selective technique for the determination of MN and SL in the presence of nine related substances that may be present in animal feed preparations. But unfortunately the proposed procedures cannot be used as stability indicating ones.

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