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Investigation of some taste substances using a set of electrodes with lipid-modified membranes

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Abstract

A five-channel taste module based on ion-selective electrodes (ISE) with lipid-polymer membranes was constructed. Five different kinds of lipids or their derivatives were used in PVC membranes as substances transforming taste information into electric signals: benzylcetyldimethylammonium chloride monohydrate, hexadecylamine, elaidic acid, 1-dodecanol and oleic acid. The electrode potential in response to sour taste substances — citric, hydrochloric and acetic acids — was examined. It was found that sensitivity of the potential of electrodes containing elaidic acid, 1-dodecanol or oleic acid in the membrane to citric and hydrochloric acid is very good. Lower sensitivity was observed in the case of acetic acid present in the system. New types of electrodes with conducting PEDT/PSS polymers and oleic acid were tested for sour taste. It was found that stability and reproducibility of electrodes used were very good. The obtained results show that lipid-polymer membranes can be successfully applied as sour taste sensors.

Keywords: Taste sensors; Ion-selective electrodes; Lipid-modified membranes; Conducting polymers

1. Introduction

In recent years many investigations were devoted to artificial sensors for use as automation of quality control in the food industry [1]. One is a multi-channel taste sensor composed of several kinds of lipid-polymer membranes [2,3]. This sensor transforms information about taste into electric signals which are then input into a computer. The image obtained shows different patterns for substances responsible for taste [1].

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Many kinds of chemical substances influence the taste of food. Human organisms do not distinguish each chemical substance [4]. Instead, humans separate the taste of food stuffs into five kinds of basic qualities: saltiness, produced mainly by sodium chloride; sourness caused by hydrogen ions (hydrochloric acid, citric acid and acetic acid); bitterness produced by caffeine, quinine or $MgCl_2$; and sweetness due to different types of sugar (sucrose, fructose, glucose) and umami produced mainly by monosodium glutamate or disodium inosinate [4].

In humans taste substances are caught by biological membranes of gustatory cells situated on the tongue, and then the information is transformed into electric signals transmitted along the nerve fibers to the brain. This biological sensor is very sensitive to taste substances, but unfortunately the response is highly subjective, depending on the nature of the organism. This is why a very selective artificial sensor is necessary in the assessment of foodstuff quality.

In this work our investigations are presented concerning stability, sensibility and reproducibility of a five-channel taste module based on ions-elective electrodes (ISE) with lipid-polymer membranes to three kinds of sour substances (hydrochloric acid, citric acid and acetic acid). Five different kinds of substances, lipids or their derivatives, were used. Also in order to improve sensitivity, stability and reproducibility of the measurements, electrodes based on lipid-polymer membranes with conducting polymer (solid-state electrodes, SSE) were used.

2. Experimental

2.1. Materials

Benzylcetyldimethylammonium chloride monohydrate, hexadecylamine, elaidic acid, 1-dodecanol, oleic acid, dioctophenyl phosphate (DOPP), and Fluka Selectrophore® reagents as high-molecular-weight PVC were used. Tetrahydrofuran (THF) distilled under $LiAlH_4$ and deionized water were used for the preparation of the solutions of taste substances. The monomer 3,4-ethylenedioxythiophene (EDOT, Baytron M) was obtained from Bayer AG and distilled under vacuum conditions before use. Poly(sodium 4-styrenesulfonate) (NaPSS, MM≈70,000) was obtained from Aldrich. The other substances used were of analytical grade.

2.2. Electrode preparation

A solid lipid-polymer membrane (ISE) was prepared in the following way: 300 mg PVC, 0.5 ml DOPP and 0.5% w/w of appropriate lipid were dissolved in THF. The solution was poured onto a glass-plate surrounded by rings and dried at 25°C in the air atmosphere. The obtained membrane film of about 200 μm thickness was transparent and colorless. The membrane was cut into 7-mm diameter discs and maintained in the electrode bodies (type IS 561) made by Moeller AG. These electrodes were filled with KCl solution (3M).

SSEs with conducting polymer were prepared by covering platinum and glassy carbon (GC) discs with thin (3 μm) films of PEDT/PSS under potentiostatic control at 0.85V vs Ag/AgCl [7]. Conducting polymer films were covered with the PVC-oleic acid membrane.

2.3. Set-up

The five-channel sensor consisted of a module of five IS-561 electrodes and a reference electrode $Ag/AgCl/Cl^-$ (3M KCl) which were immersed in the appropriate electrolyte of the taste solution. The electrode potentials of each channel were measured separately using a high-input impedance voltmeter (Hach EC30) (Fig. 1). The electrodes were conditioned in 0.1 M KCl prior and between measurements. All the experiments were carried out at a temperature of 23± 0.5°C. All SSE electrodes were tested separately as a one-channel probe.

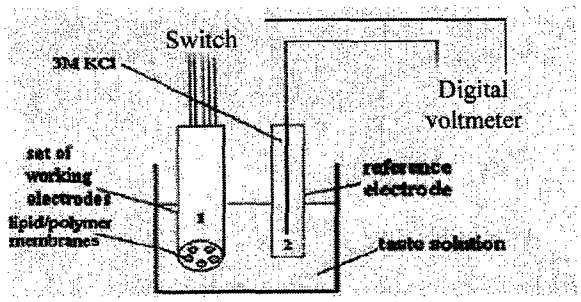

Fig. 1. Experimental set-up: 1 five channel sensor, 2 Ag/AgCl/Cl^- reference electrode.

The following lipids and lipid derivatives were used to transform taste information into electric signals: benzylcetyldimethylammonium chloride monohydrate, hexadecylamine, elaidic acid, 1-dodecanol, oleic acid. All SSEs were tested for oleic acid.

3. Results and discussion

The influence of the concentration of sour substances (hydrochloric, citric and acetic acids) on the ISE electrode containing hexadecyamine is presented in Fig. 2. As can be seen, the electrode potential, E, has changed significantly with the rise of citric acid (Fig. 2, curve b) and hydrochloric acid (Fig. 2, curve a) concentration in the range of 10^{-3}–1 M. In the case of acetic acid (Fig. 2, curve c), significant changes of E were observed only for its high concentrations (1–0.1 M). This system is not sensitive to concentrations of these sour substances below 10^{-3} M.

The situation improves when benzylcetyldimethylammonium chloride monohydrate (Fig. 3) is used instead of hexadecylamine. Electrode potential changed significantly even with a very low concentration of citric acid (Fig. 3, curve b) and hydrochloric acid (Fig. 3, curve a). Again, this is not the case for acetic acid (Fig. 3, curve c), when the electrode potential is barely sensitive to its concentration up to 0.1 M.

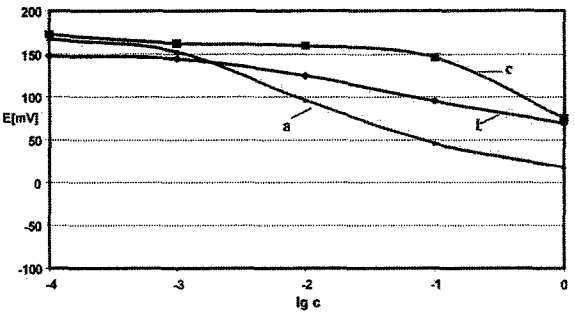

Fig. 2. Potential of the electrode with hexadecylamine in the membrane as a function of the concentration of: a, hydrochloric acid; b, citric acid; c, acetic acid.

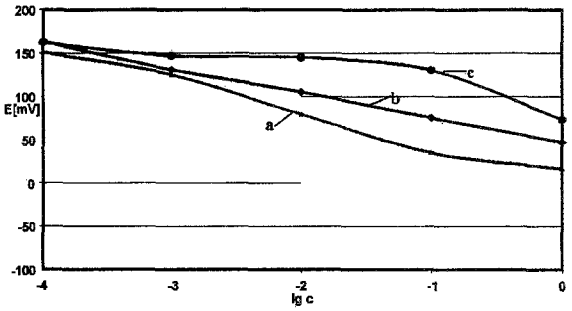

Fig. 3. Potential of the electrode with benzylcetyldimethylammonium chloride monohydrate in membrane as a function of the concentration of: a, hydrochloric acid; b, citric acid; c, acetic acid.

Completely different $E = f(\log c)$ dependencies were obtained for the three other systems containing 1-dodecanol, elaidic acid and oleic acid, respectively (Figs. 4–6). For small concentration (10^{-4} M) the potential has a value of about −70 mV. This was maintained for all the acid nature systems investigated independently of the three lipid (1-dodecanol, elaidic acid, oleic acid) membranes used (Figs. 4–6). This potential visibly increased with the rise of concentration of the hydrochloric (curve a) and citric acid (curve b). Again, as in previously described systems (Figs. 2 and 3), a smaller sensitivity to the amount of acetic acid molecules (curve c) was observed.

Summarizing, we can see that the potential of electrodes with 1-dodecanol, oleic acid and

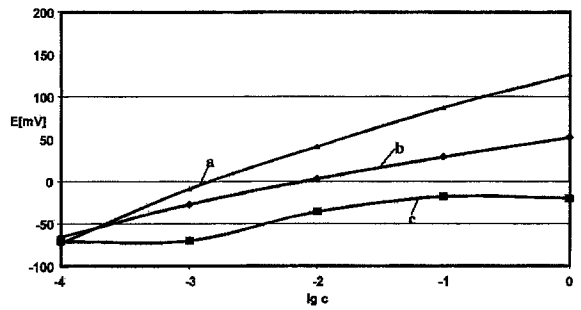

Fig. 4. Potential of the electrode with 1-dodecanol in the membrane as a function of the concentration of: a, hydrochloric acid; b, citric acid; c, acetic acid.

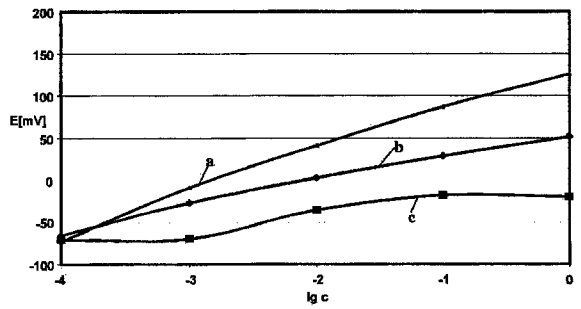

Fig. 5. Potential of the electrode with elaidic acid in the membrane as a function of the concentration of a, hydrochloric acid; b, citric acid; c, acetic acid.

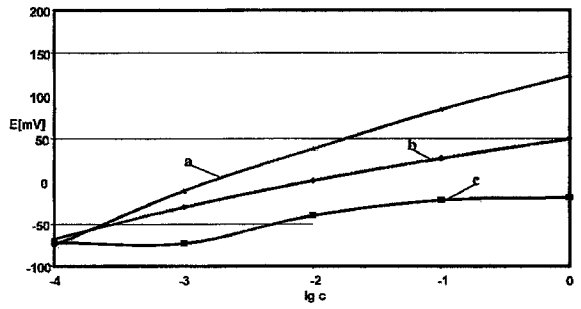

Fig. 6. Potential of the electrode with oleic acid in the membrane as a function of the concentration of: a, hydrochloric acid; b, citric acid; c, acetic acid.

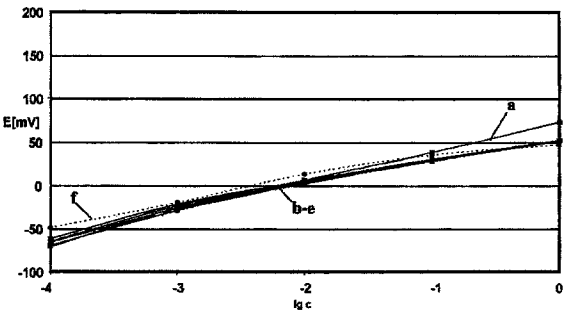

Fig. 7. Stability for the elaidic acid polymer membrane after: a, 1 day; b, 2 days; c, 3 days; d, 4 days; e, 5 days; f, 21 days.

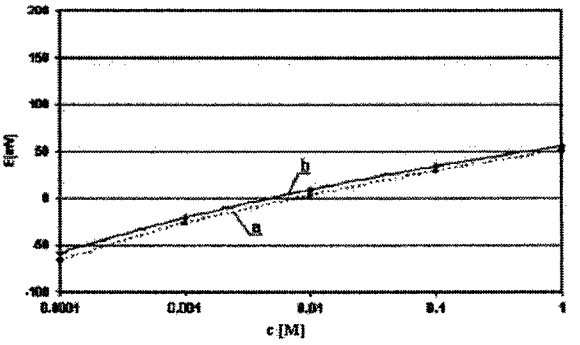

Fig. 8. Reproducibility of system containing the elaidic acid polymer membrane for citric acid: a, first preparation; b, second preparation.

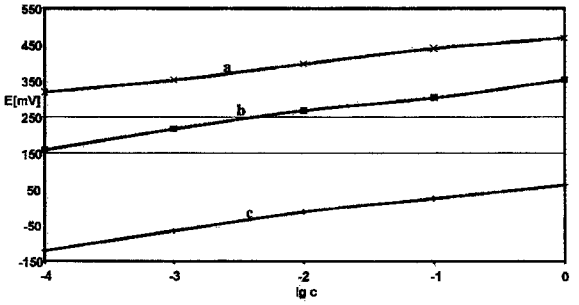

Fig. 9. Dependence $E = f(c)$ for three types of electrodes: a, SSE/Pt; b, SSE/GC; c, ISE. Lipid material, oleic acid; taste substance, HCl; reference electrode, $Hg/Hg_2Cl_2/Cl^-$ (3 M KCl).

elaidic acid are very sensitive to sourness caused by citric or hydrochloric acid and much less to acetic acid. However, it should be noted that in the range of concentration between 10^{-3} M and 10^{-2} M, where the mean human sour taste threshold is contained ($2*10^{-3}$ M) [5], the sensitivity of a set of three electrodes is quite good.

In order to use these systems as potentiometric taste sensors for sourness, the stability of lipid-polymer electrodes should be investigated. The appropriate results for the system containing elaidic acid as a lipid membrane are presented in Fig. 7. It can be concluded that the electrode stability during a period of 4 days is very good. After the fifth day potential changes in the range of high concentrations. The opposite effect, higher potential changes in the range of low concentration, was observed after 21 days. Similar or even better stability was noted for systems containing other lipids.

In order to use such a system as a taste sensor, the reproducibility of the results concerning newly prepared membranes should be known. Appropriate investigations for the system containing elaidic acid are presented in Fig. 8. As can be observed, the reproducibility of the results with newly prepared membrane is very good.

To improve measurements and simplify sensor construction, electrodes based on lipid-polymer membranes with conducting polymer PEDT/PSS SSEs were also examined [6]. As can be seen in Fig. 9, the slope of the $E = f(\log c)$ curves is the same, showing a direct relationship between sour substance concentration and the sensor response. The slope $\Delta E/\Delta \log c$ is equal to 37 mV, 39 mV and 40 mV for SSE/Pt, SSE/GC and ISE electrodes, respectively. All solid-state electrodes exhibited good stability over a long time period (about 2 weeks). The conducting polymer used in the electrode allows reducing the number of elements in the electrode sensor. This type of electrode does not require an inner solution. The conducting polymer film plays the role of ionic and electronic conductor and is a stable source of redox potential. Our preliminary results prove that the desired simplifications of electrode construction do not deteriorate the sensitivity of the membrane response.

In conclusion, it may be stated that this new set of channels could be successfully applied in the estimation of the sourness in food.

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