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Multichannel taste sensors with lipid, lipid like – polymer membranes

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Abstract. The elaboration of a sensitive taste sensor for discrimination of different soft drinks is very important in food industry. The short review of taste sensors described in the literature is presented. Two types of potentiometric taste sensors, one with lipophilic compound-polymer membranes (ISE) and the other with lipid polymer membrane and a conducting polymer film (All solid state electrode, ASSE) were tested in appropriate taste solutions. Five channel ISE sensor was examined in acid, sour and sweet solutions. This sensor was sensitive to bitter and sour substances and not too sensitive to sucrose concentration. It was successfully used for discrimination of different kind of soft drinks. Four channel ASSE sensor was examined in sour solutions. It was found that stability and sensitivity of ASSE are lower than ISE. Therefore, it seems that the previous one cannot be applied in taste sensor.

There are five kinds of basic qualities of taste: saltiness, produced mainly by sodium chloride, sourness caused by hydrogen ions of acids (hydrochloric, acetic or citric), bitterness produced by quinine hydrochloride or caffeine, sweetness due to different types of sugars (e.g. sucrose, fructose) and umami produced mainly by monosodium glutamate [1]. In fact, the taste of food products is developed by many substances which affect each other. In general, taste is estimated by organoleptic methods. In human organisms taste molecules are caught by biological membranes of gustatory cells situated on the tongue and the information is transformed into electric signal which is transmitted by the nerve fibers to the brain. Such biological sensor is very sensitive to taste substances, but unfortunately the response is subjective, depending on the nature of the vivid organism. Therefore, a lot of investigations have been focused on elaboration of an artificial taste sensor for quality control of food products or recognition of similar products form different branches [2-12].

Multichannel taste sensor based on potentiometry, developed at Kyushu University (Japan) was composed of eight different lipid-polymer membranes [2]. The potential difference between the electrodes and reference electrode was measured by milivoltmeter and collected by computer. The electrodes play the role of transducer transforming taste information generated by chemical substances into electrical signals. This sensor was successfully used in laboratory conditions for discrimination of several liquid products as beer, coffee, wine, milk, tomatoes and mineral water [2-4,12]. However, despite many investigations this sensor still is not applied for larger scale.

The other taste sensor developed at Linkoping University (Sweden) was based on voltammetry, and made from four or five wires of different metals (gold, iridium, palladium, platinum and rhodium), a reference electrode and an auxiliary electrode. The measurement principle is based on pulse voltammetry in which current is measured during the change of the amplitude of the applied potential. Its application was not as wide as in the case of previous sensor, but it was successfully used for monitoring of freshness of milk, [5] quality of orange juice [6] and for discrimination of different teas and solutions of detergents [7]. Three different electrodes, traditional Pt and Au electrodes and the third one modified with poly(3,4-ethylenedioxythiophene) (PEDOT), conducting polymer, were tested in Modena University (Italy) for discrimination of different fruit juices [11]. It was shown that the electrode modified with conducting polymer demonstrated the most discriminating ability.

1

The comparison of the responses of potentiometric and voltammetric sensors were done using multivariate data analysis based on principal component analysis [7]. It was shown that extra information can be gained by combination of the two sensor systems.

The other potentiometric taste sensor (figure 1) made from five electrodes containing lipophilic compound-polymer membranes and reference electrode has been proposed at Gdańsk University of Technology (Poland) [8-10]. Five lipophilic compounds (benzylhexadecyldimethylammonium chloride monohydrate, hexadecylamine, elaidic acid, 1-dodecanol, cholesterol) were applied in PVC with plasticizer (dioctyl phenylphosphonate) matrix forming five polymeric membranes.

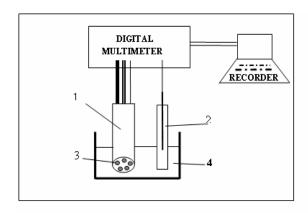


Figure 1. Lipid, lipid like-polymer taste sensor: 1 – set of working electrodes filled with 0.1 KCl, 2 – Ag/AgCl/Cl- reference electrode, 3-lipid, lipid like-polymer membranes, 4- taste substances.

This sensor was tested in sour solutions (hydrochloric, acetic and citric acids) [8]. It was found (figure 2) that the response of electrodes containing positively charged membranes (benzylhexadecyldimethylammonium chloride monohydrate or hexadecylamine) decreases with increasing of sour substance concentration, meanwhile the effect is opposite in case of negatively charged membranes (elaidic acid, 1-dodecanol or cholesterol). Moreover, the latter membranes exhibited much higher diffusion resistance in comparison to positively charged membranes [10].

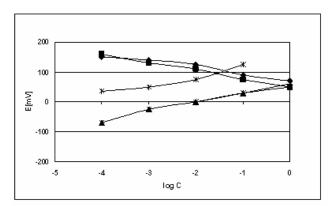


Figure 2. Potential changes of electrodes with positively charged membranes: benzylcetildimethylammonium chloride monohydrate (**a**), hexadecylamine (**4**) and negatively charged membranes: elaidic acid (**A**), 1-dodecanol (x), cholesterol (*) with citric acid concentration.

This sensor was also tested in sweet and bitter solutions. The results are presented in figure 3 a, b.

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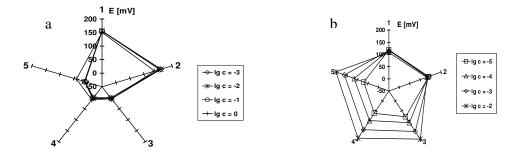


Figure 3. Changes of electric potential of five membrane electrodes containing: benzyldecyldimethylamonnium chloride monohydrate (1), hexadecylamine (2), 1-dodecanol (3), elaidic acid (4), cholesterol (5) with the concentration of sucrose (a) or quinine hydrochloride (b) [9].

It should be concluded from figure 3a that all the membrane electrodes were almost not sensitive to sucrose concentration. However, the responses of two membrane electrodes (no 2 and 5) differ in very high sucrose concentration [9]. In the case of bitter substances, three electrodes with negatively charged membranes were very sensitive to quinine hydrochloride (3, 4 and 5 in figure 3b). On the contrary, the positively charged membranes (1 and 2 in figure 3b) were not sensitive to bitter substance concentration changes.

The mechanism of lipid-polymer membrane response is not fully recognized [12]. From electrochemical impedance spectroscopy measurements [10] it was found that negatively charged membranes exhibit higher diffusion resistance in comparison to positively charged membranes. Therefore, the membrane potentials of the negatively charged membranes originate mainly from surface potential, whereas both the surface and diffusion potentials play the role in positively charged membranes. It is believed that lipid molecules are packed densely in the membrane [12]. The degree of binding of a taste substance with the lipid molecules in the membrane depends on hydrophobic and electrostatic interactions and the balance between hydrophilicity and hydrophobicity of the taste substance.

The five-channel taste sensor with lipophilic compound-polymer membranes (ISE) was successfully applied for discrimination and quality control of different kinds of soft drinks containing sucrose, quinine hydrochloride and carbon dioxide (tonics from different brands) [9]. The electrode responses were transformed by multivariate analysis (PCA) (figure 4) and compared with test performed by organoleptic methods. It was found that the tested soft drinks can be classified into three main groups, each of different taste. The taste within the same group was similar. The taste of the drink Nata (no 2) differs significantly from the other ones, therefore the point representing this drink is outside these three groups.

From the results presented above it seems that five-channel taste sensor with lipophilic compoundpolymer membranes could be successfully applied for discrimination of different kind of soft drinks composed of sugar, citric acid, quinine hydrochloride and carbon dioxide.

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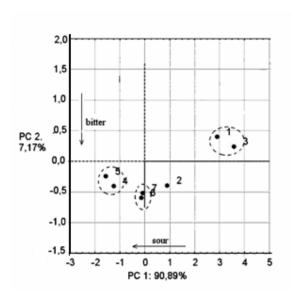


Figure 4. PCA plot for the first two principal components of data obtained for eight brands of tonics: 1 – Jurajski, 2 – Nata, 3 – Hellena, 4 – Schweppes Indian, 5 – Kinley, 6 – Kinley Lemon, 7 – Schweppes Bitter Lemon.

To improve measurements and simplify sensor construction, ASSE based on lipid-polymer membranes with conducting polymer PEDOT/PSS were proposed as taste sensor [13]. The advantage of ASSE in comparison to ISE is that the former ones are the solid state electrodes.

ASSE were prepared by coating GC/PEDOT electrodes with an ion selective lipophilic compounds-polymer membrane (mass ratio: lipophilic compound/PVC = 0.033). The thickness of conducting polymer and lipophilic compound-polymer membrane was $1.0\mu m$ and $10\mu m$, respectively.

The four membrane electrodes (with benzyldimethyltetradecylammonium chloride (1), dodecyltrimethyl ammonium bromide (2), palmitic acid (3), stearic acid (4) responses in different citric acid concentration are presented in figure 5.

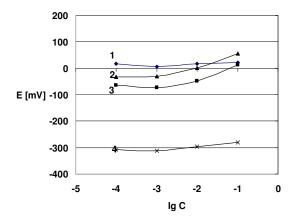


Figure 5. Electric potential changes of ASSE with \bullet - benzyldimethyltetradecylammonium chloride (1), \blacktriangle - dodecyltrimethyl ammonium bromide (2), \blacksquare - palmitic acid (3), \mathbf{x} - stearic acid (4) as a function of citric acid concentration [13].

It is shown that ASSE electrodes are not too sensitive to citric acid concentration. Moreover, for both types of membrane electrodes: positively charged (benzyldimethyltetradecylammonium chloride,

dodecyltrimethyl ammonium bromide) and negatively charged (palmitic acid and stearic acid) potential values increase with citric acid concentration. This observation is different in case of ISE electrodes.

The stability of ISE and ASSE electrodes was also examined. The results obtained within eight days for ASSE with stearic acid and ISE with elaidic acid in different citric acid concentrations are presented in figure 6. It is seen that ISE with elaidic acid is more stable than ASSE with stearic acid. Similar behaviour was found for other ISE and ASSE in citric acid solutions. Therefore, ISE seem to be better for application in taste sensor, taking into account the stability and sensitivity of two types of electrodes used.

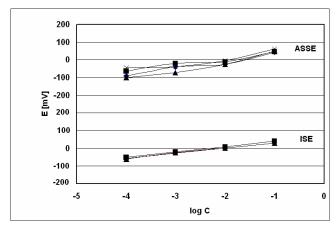


Figure 6. Stability of ASSE and ISE as a function of citric acid concentration after \blacktriangle - 1 day, \blacksquare - 2 days, \blacklozenge - 3 days, x - 8 days, ASSE with stearic acid, ISE with elaidic acid.

Summarizing, five-channel sensor containing ISE electrodes is more stable and more sensitive than four-channel sensor with ASSE electrodes containing the conducting polymer layer. The previous one can be successfully applied for discrimination of different kind of soft drinks.

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