

Application of a potentiometric electronic tongue as a classification tool in food analysis

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Abstract

This paper reports on the application of a potentiometric sensor array to the food analysis field, in order to distinguish simple tastes and to classify food samples. This array is formed by a set of non-specific all-solid-state potentiometric sensors and has been used in combination with principal component analysis (PCA) for the classification of food samples in batch and in flow injection mode. First attempt was to classify synthetic samples prepared with controlled variability. Once this ability is proven, satisfactory classification results are presented for commercial waters, orange-based drinks and tea samples. An interesting correlation is achieved between the natural juice content and its first calculated component, which allows for a very simple tool for screening purposes.

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1. Introduction

Recent trends in the analytical chemistry field, take direction opposed to those that invest efforts in the development of quasi-specific instruments. The new approach departs from the use of general-purpose devices, combined with computer processing stages, which could probably grasp looks or composition of a sample and transmit us information about it [1].

These new strategies are based on the use of sensor arrays with a generic cross response to a wide spectrum of analytes; with those, we can obtain different aspects of information about the sample. Afterwards, the use of multivariate calibration tools is used to extract the sought results from the data generated by the devices [2].

This strategy is already known, for analyte detection on aqueous samples, as electronic tongue, or for gases or headspace samples, as electronic nose. The used terminology suggests that this approximation is bioinspired on animal taste or olfaction senses, where a few receptors can respond to a large variety of substances. With taste, only generic infor-

mation is used, like sweet, bitter, salted or acid; additionally, this approach entails an advanced data treatment mechanism next applied by the brain, in order finally to recognize, quantify or classify between different substances.

An interesting application of this new concept is the use of chemometric tools for multivariate calibration and multiple analyte recognition simultaneously, which is a good strategy to multiparametric determination without interference removal [3–5]. A second interesting application is their use inspired in the human brain to classify the nature of a sample. For this purpose, principal component analysis (PCA) is employed as data treatment. Significant work has been performed in this field, specially, for gas sensors, or electronic noses [6–10].

The use of non-selective potentiometric sensors arrays in combination with PCA starts to be an analytical alternative for sample classification or identification, especially in food analysis [11]. Various contributions on this topic can be found in the literature. Per example, Toko used an electronic tongue to classify and assess the quality of various drinks including wine and water [12]. Wróblewski et al. were able to distinguish between different brands of beverages using a similar approximation [13,14]. Apart, Krantz-Rülcker et al. moni-

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tored a mineral water bottling plant with a voltammetric electronic tongue [15]. Few references are, however, in respect to the use of an e-tongue for quantitative purposes; such references include the determination of various species in water and wine [16], or beer and soft drinks [17,18].

In this work, we used a potentiometric sensor array formed by generic and selective devices presenting response to other interfering species. Each sensor used neutral carriers in a polymeric PVC membrane cast on a solid inner contact, and PCA is employed as the processing tool. First, we set the device to classify manually prepared samples with certain amount of variability. Once the classification ability is proven in controlled experiments, we applied the electronic tongue in food analysis to classify various types of commercial waters, some orange-based drinks and, finally, some teas.

2. Experimental

2.1. Chemicals

The ion-selective poly(vinyl chloride) (PVC) membranes were prepared from high-molecular weight PVC (Fluka), using *bis*(1-butylpentyl)adipate (BPA), dioctylsebacate (DOS), *o*-nitrophenyloctyl ether (*o*NPOE), dioctylphenylphosphate (DOPP), tris-ethylhexyl phosphate (TEHP) and dibutylsebacate (DBS) (all from Fluka) as plasticizers. The ionophores employed to formulate the potentiometric membranes were nonactin (ammonium ionophore I, Fluka), valinomycin (potassium ionophore III, Fluka), 2,3:11,12-didecalino(16-crown-5) (ionophore DD16C5, Dojindo), lasalocid (Fluka), dibenzo(18-crown-6) (Fluka), tridodecylamine (Fluka), 2,9-di-*n*-butyl-1,10-phenantroline (TCI) and bis(bis(4-1,1,3,3-tetramethylbutyl)phenyl)phosphato calcium (II) (BBTP, Fluka). Potassium tetrakis(*p*-chlorophenyl)borate (Fluka) was used, when necessary for a correct potentiometric response.

The materials used to prepare the solid electrical contact were the epoxy resin components Araldite M and Hardener HR (both from Ciba-Geigy), and graphite powder (100 μm , BDH) as conductive filler.

Imidazole (Fluka), Tris base, and the salts NH_4Cl , KCl , NaCl , LiCl and CaCl_2 (reagent-grade, Merck), were used as background electrolytes and calibration species, respectively.

All solutions were made in deionised, highly purified water (16–18 $\text{M}\Omega\text{ cm}$ resistivity, Milli Q, Millipore).

2.2. Sensor array

The sensors were all-solid-state ion selective electrodes (ISEs) with a solid electrical contact made from conductive composite. They were constructed by following standard procedures developed from our laboratory [4,19]. Each device was constructed from a PVC cylinder of 100 mm length and 6 mm internal diameter. Polymeric PVC membranes were formed by solvent casting of a mixture further diluted with tetrahydrofuran (1 ml per 20 mg PVC) on a 1:1 epoxy-graphite composite solid contact. Membranes were obtained by eight cumulative-additions followed by evaporation of 50 μL drops of each cocktail membrane. Once formed, membranes were conditioned in a 0.1 M solution of their primary ion for 24 h. Arbitrarily, for the generic electrodes, we used ammonium ion for its conditioning. Each device was constructed by triplicate and was previously evaluated to check if presented correct response characteristics. The specific formulation of the different membranes is detailed in Table 1.

2.3. Apparatus

Potentiometric measurements were performed with a laboratory-made data acquisition system consisting of 32 input channels made with differential instrumentation amplifiers (INA116, Burr-Brown, USA) that adapted the impedance for each sensor. Measurements were referred to an Ag/AgCl reference electrode (Thermo Orion 90-02-00). Each channel was noise-shielded with its signal guard. The output of each amplified channel was filtered with a second order low pass active filter centered at a 2 Hz frequency and connected to an Advantech PC-Lab 813 A/D conversion card installed in a PC. Readings were acquired by using custom software developed by the authors in Microsoft QuickBasic Version 4.5.

Potentiometric measurements were made by using solutions containing 0.010 M imidazole buffer at pH 6.50 and Tris at pH 7.5, 8.0 and 8.5 as background media depending on each application.

Table 1
Compositions of the used potentiometric sensors, percentages are by weight, unless specified

Electrode	Ionophore	PVC (%)	Plasticizer
H^+ (1)	Tridodecylamine 1% (10% molar potassium tetrakis(<i>p</i> -chlorophenyl)borate (KTpCIPB))	33	DOS 66%
Li^+	2,9-Di- <i>n</i> -butyl-1,10-phenantroline 1.4% (66% molar KTpCIPB)	27.8	<i>o</i> NPOE 69.8%
NH_4^+	Nonactin 1%	33	BPA 66%
K^+	Valinomycin 3%	30	DOS 67%
Na^+	DD16C5 3% (10% molar KTpCIPB)	29.1	TEHP 67.9%
Ca^{2+}	BBTP 4.3%	30.3	DOPP 65.4%
GENERIC 1	Dibenzo(18-crown-6) 4%	29	DOS 67%
GENERIC 2	Lasalocid 3%	27	DBS 70%
H^+ (2)	Tridodecylamine 1% (10% molar KTpCIPB)	33	DBS 66%

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