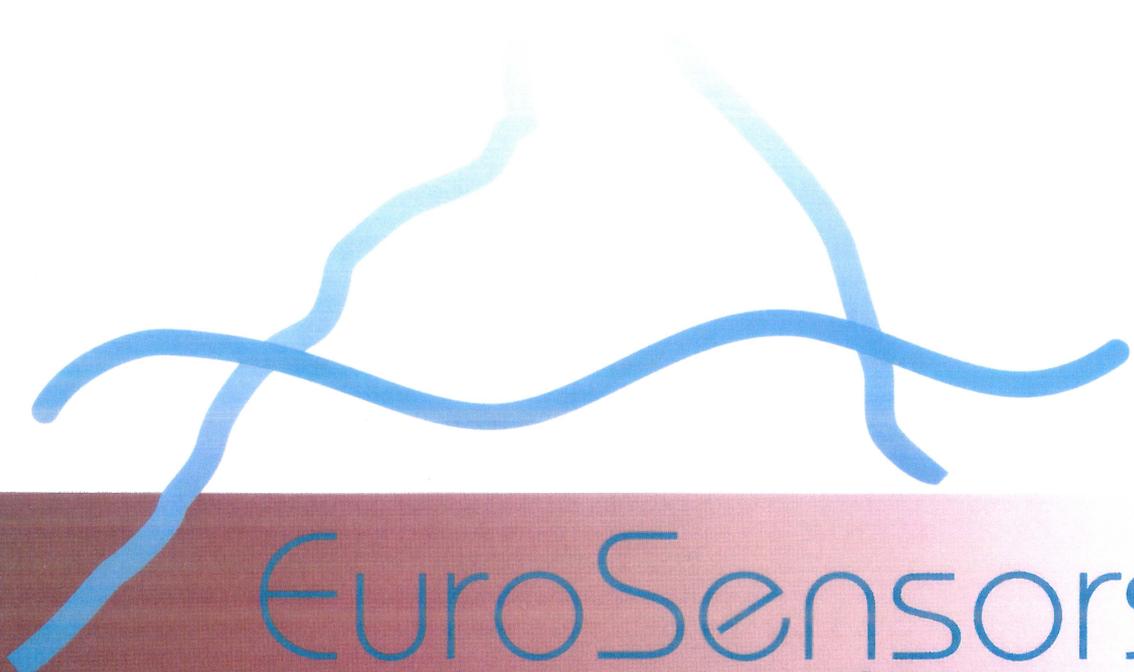


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An electronic tongue for juice level evaluation in non-alcoholic beverages

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Summary

An electronic tongue with 36 cross-sensitivity polymeric membranes was built and applied for qualitative beverage analysis. The objective was to distinguish non-alcoholic beverages with different levels of added fruit juice. The different signal profiles recorded by the electronic tongue device, together with a supervised multivariate statistical method for pattern recognition, were used to differentiate 4 beverage groups with different added fruit juice contents: higher than 30%, between 14% and 30%, between 5% and 10% and between 0.1% and 2%. A set of 16 portuguese beverages (4 for each group), purchased in commercial supermarkets, were analyzed. A stepwise linear discriminant analysis was used allowing a very satisfactory overall correct classification of 100% for the original grouped cases and of 93.8% for the "leaving one-out" cross-validation procedure.

Motivations and results

In the last decades, several works have reported the application of electronic tongue (e-tongues) devices for distinguishing among various types of foods (beer, coffee, honey, juice, milk, mineral water, wine) [1-11]. The increasing interest regarding e-tongue application in food area can be partially attributed to the lower calibration costs, satisfactory accuracy for reasonable small sizes of the calibration data set and easy adaptability to different working conditions, when compared with other analytical methodologies [3]. However, in literature works, namely those regarding the study of juices [7-11], the e-tongue devices were applied mainly to recognize and differentiate juice brands. In this work a multi-sensor array device was used for non-alcoholic beverages discrimination according to the juice content.

The e-tongue device built consisted of 36 polymeric membranes divided in two-sensor array (Figure 1). Each membrane was prepared using PVC, as polymeric matrix, different plasticizer compounds and membrane additives (Table 1). The multi-sensor system, together with a double junction Ag/AgCl reference electrode, was connected to a multiplexer Agilent Data Acquisition/Switch Unit model 34970A. Measurements were performed in a double wall glass cell thermostated at 25 °C. All beverages were analysed during a 10-15 minutes period.

The ability of the multi-sensor device to recognise beverages with 4 different added fruit juice levels was evaluated using 16 commercial beverages of different brands. Each group consisted in 4 beverages of the same brand with different compositions and with different kind of added fruit juices within groups. The 4 groups of beverages considered were: A (juice level higher than 30%), B (juice level between 14% and 30%), C (juice level between 5% and 10%) and D (juice level between 0.1% and 2%).

The multi-sensor signal pattern recorded for the beverages analysed (samples were analyzed twice), together with a stepwise linear discriminant analysis (LDA), was used to distinguish the 4 beverages groups. The stepwise LDA technique selected only the signals of 4 polymeric membranes for the final model ($p < 0.001$ for Wilks' Lambda test). Two significant discriminant functions were established ($p < 0.001$) accounting for 99.0% of the total variance. In Figure 2 the scores for the two functions were plotted (explaining 78.4% and 20.6% of the total variance, respectively). As shown in the figure, the approach had a very satisfactory performance allowing 100.0% and 93.8% correct classification for original grouped cases and "leaving one-out" cross-validation procedure, respectively.

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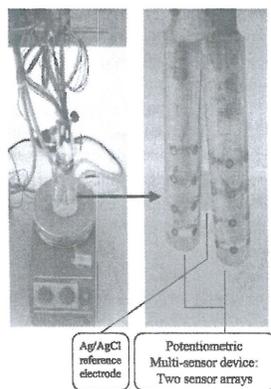


Fig. 1: E-tongue device

Table 1 - Additives and plasticizers used in the polymeric membranes preparation

| Membrane Additive substance | Plasticizer substance |
|------------------------------------|-----------------------------|
| Octadecylamine | Bis(2-ethylhexyl)phthalate |
| Bis(2-ethylhexyl)phosphate | Bis(1-butylpentyl) adipate |
| Oleyl alcohol | Tris(2-ethylhexyl)phosphate |
| Methyltriocetylammmonium chloride | Dibutyl sebacate |
| Tridodecylmethylammmonium chloride | 2-Nitrophenyl-octylether |
| Oleic acid | Diocetyl phenylphosphonate |

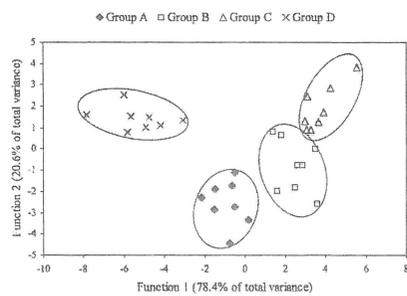


Fig. 2: Discriminant analysis obtained for the juices recognition.

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INTRODUCTION

An electronic tongue with 36 cross-sensitivity polymeric membranes was built and used for semi-quantitative analysis of beverages.

The multi-sensor device was used to recognise different added fruit juice levels in commercial beverages of different brands. Four beverages groups were considered (Table 1):

(A) fruit drinks with addition of fruit juice above 30%;

(B) fruit drinks with added fruit juice between 14% and 30%;

(C) gasified juices with a percentage of fruit juice between 6% and 10%;

(D) ice tea drinks with addition of less than 4% of fruit juice.

Table 1. Description of the beverages analyzed

| Group | Beverage | Composition as provided in label |
|-------|--------------------------------------|-------------------------------------|
| A | Pineapple juice "Brand I" | >50% of juice |
| | Mango juice "Brand I" | >30% of juice |
| | Peach juice "Brand I" | >50% of juice |
| | Tutti-Frutti juice "Brand I" | >50% of juice |
| B | Orange juice "Brand II" | 20% of juice |
| | Mix fruits juice "Brand II" | >20% of juice |
| | Pineapple juice "Brand II" | >20% of juice |
| | Strawberry juice "Brand II" | >14% of juice |
| C | Pineapple juice with gas "Brand III" | 6% of juice |
| | Orange juice with gas "Brand III" | 8% of juice |
| | Pineapple juice with gas "Brand IV" | 8% of juice and pulp |
| | Orange juice with gas "Brand IV" | 10% of juice and pulp |
| D | Mango ice tea "Brand V" | 0.13% of juice and 4.7% of tea |
| | Lemon ice tea "Brand V" | 0.1% of juice and 4.7% of tea |
| | Peach ice tea "Brand V" | 0.1% of juice and 4.7% of black tea |
| | Red ice tea "Brand V" | 3.4% of juice and 3.3% tea extract |

POTENTIOMETRIC ELECTRONIC TONGUE

The e-tongue device consisted of 36 polymeric membranes applied into two-sensor arrays, together with a double junction Ag/AgCl reference electrode (Figure 1).

The device was connected to a multiplexer Agilent Data Acquisition/Switch Unit model 34970A. Measurements were performed in a double wall glass cell thermostated at 25 °C (Figure 2).



Figure 2. Multi-sensor system:

1- PC for data acquisition; 2- DataLogger Agilent; 3- E-tongue device; 4- Magnetic stirrer; 5- Thermostatic bath.



Potentiometric Multi-sensor device: Two sensor arrays
Ag/AgCl Reference electrode

Figure 1. E-tongue device.

Table 2. Additives and plasticizers used for polymeric membranes preparation

| Membrane Additive substance | Plasticizer substance |
|---------------------------------------|---------------------------------|
| [1] Octadecylamine | [A] Bis(2-ethylhexyl)phthalate |
| [2] Bis(2-ethylhexyl)phosphate | [B] Bis(1-butylpentyl) adipate |
| [3] Oleyl alcohol | [C] Tris(2-ethylhexyl)phosphate |
| [4] Methyltriocylammonium chloride | [D] Dibutyl sebacate |
| [5] Tridodecylmethylammonium chloride | [E] 2-Nitrophenyl-octylether |
| [6] Oleic acid | [F] Dioctyl phenylphosphonate |

DISCRIMINANT ANALYSIS

A linear discriminant analysis (LDA), with stepwise technique, was used to classify the non-alcoholic beverages according to their juice contents using the e-tongue recorded signal patterns.

A leaving-one-out cross-validation procedure was carried out to assess the model performance.

The LDA statistical analysis was performed at a 5% significance level using the SPSS software, version 17.0 (SPSS Inc).

SAMPLE ANALYSIS

The 16 beverages were analysed twice, without any dilution, during a 10-15 minutes period. Gasified juices were previously degassed using an ultrasound device during 3min.

RESULTS

A classification model based on the signals of 4 polymeric membranes (A4, D5, E1 and F5; $P < 0.001$, Wilks' Lambda test) was obtained.

Two canonical discriminant functions ($P < 0.001$, Wilks' Lambda test) were selected explaining 99.0% of the total experimental data variance.

The approach had a very satisfactory performance, allowing a 100.0% and 93.8% correct classification for the original grouped cases and the "leaving one-out" cross-validation procedure, respectively (Figure 3).

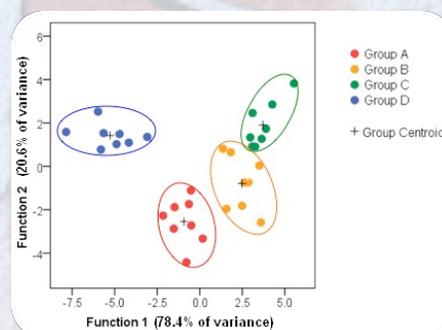


Figure 3. Discriminant analysis obtained for the juices recognition.

CONCLUSIONS

For the first time, to the best of the authors' knowledge, an e-tongue device was successfully applied, directly on real samples, for juice semi-quantitative discrimination, based on the fruit content range level.

Each membrane was prepared using (Table 2):

31.9-32.3% of PVC;

64.7-65.2% of one of the plasticizers;

2.8-3.2% of one of the membrane additives.

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