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Description and characterisation of a large array of sensors mimicking an artificial olfactory epithelium

Mara Bernabei, Simone Pantalei, Krishna C. Persaud*

School of Chemical Engineering and Analytical Science, The University of Manchester, Manchester, M13 9PL, UK

Abstract

Biological olfactory systems show high sensitivity and exquisite discriminatory capacity to odorants. These characteristics are due to hierarchical signal processing of the large numbers of sensory inputs that occurs within the olfactory system. In testing realistic computational models of the olfactory system, large numbers of chemical sensor inputs are required. So far, sensory devices that may serve as model inputs to an artificial olfactory system do not exist. The development of a large scale array of chemical sensors able to mimic the olfactory receptor neurons is described, and these have been characterised in terms of their variability and degree of redundancy. Using this device it is possible to start testing computational hypotheses appropriate to biological chemosensory systems and adapt them to the artificial olfaction.

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Keywords: Biomimetic olfaction, large sensor array, conducting polymer sensors

1. Introduction

The unique structure of the biological olfactory system allows detection and discrimination of a wide range of odours, giving vital information on food, predator, mates and pathogens [1]. While there has always been a great interest in developing instruments that mimic olfaction, so far, no apparatus having the same performance as biological olfaction exists. Understanding of the olfactory structure [2-7] and coding mechanisms have stimulated new bioinspired computational solutions for signal processing. However, without realistic sensor data inputs such

* Corresponding author. Tel.: +44-(0) 161 3064892.
E-mail address: Krishna.persaud@manchester.ac.uk
models can neither be benchmarked nor completely exploited. Our ideas were to develop a large sensor array that, mimicking the olfactory epithelium (OE), can be interfaced with computational models of the biological olfactory system to provide them realistic sensory inputs, allowing the evaluation the performance of these bioinspired models. Hence, we built a modular instrument capable of acquiring data from 65536 sensors by reading in parallel 16 sensor arrays. Each array has 4096 elements arranged in matrix configuration. Interdigitated transducers with different geometries were implemented in the array to increase the sensor dynamic range variability. Conducting polymers were chosen as sensing materials as their characteristics were considered suitable for our aim. Presently, the system has 16384 sensors consisting of 25 different types. Detailed description of the instrument architecture can be found in [8,9].

Laboratory experiments have been performed to assess that the array has the desired features to mimic the OE. Here we report characterization of variability and redundancy of sensors in the synthetic array.

2. Methods

2.1. Artificial olfactory epithelium: features and architecture

The OE is able to detect a large variety of odorant molecules with a unique level of sensitivity, due to a very large number of olfactory receptor neurons (ORNs) exhibiting broad and overlapping specificity and characterized by a high level of redundancy. The dynamic responses of the ORNs are also important for odour discrimination. Different ORNs exhibit different dynamic activity when reacting to a stimulus and, moreover, receptors of the same type often show significant differences in responses to different concentrations of the same odorant. This variability may add significant information content to the patterns of responses generated across the entire olfactory mucosa [10-12].

To imitate these important properties, we developed a modular system able to read 16 sensor arrays in parallel. Each array is composed of 4096 polymer sensors, so the whole device is able to read up to 65536 sensors. Moreover, the modular approach allows the number of sensors that can be implemented in the instrument to be increased easily, by replicating the electronics developed. Besides a large number of sensors, we wanted also to provide the system with high level of redundancy and sensor dynamic variability similar to that of the receptors. This was achieved by using sensing materials based on conducting polymers and deposited on transducer electrodes with different geometries.

Conducting polymers have a backbone consisting of alternating single and double bonds, which leads to the formation of π-electrons partially delocalised across few atoms of the polymer system. The delocalised states cause the formation of a band gap (Eg). The Eg of the most conducting polymers is quite large and undoped polymers are practically insulating. A doping process performed by redox reactions or protonation, increases their electrical conductivity. A secondary dopant can interact with a primary-doped polymer, inducing changes of its physical properties due, principally, to a variation of the molecular conformation [13,14]. The secondary process is important in the sensing field as many analyte gases act as secondary dopant for conducting polymers, making them exploitable as sensing materials [14]. In addition, the affinity of a polymer for a secondary dopant can be tuned by using post treatment or creating specific sites for binding the molecules [15], so that numerous different sensing polymeric materials are feasible. We focused on three families: polyanilines, polypyrroles and polythiophenes. Their sensitivities were differentiated by using a variety of dopant species, adding functional group to the main polymer ring and preparing copolymers. This resulted in 25 polymers listed in Table 1.

In order to increase the variability within the sensors and to introduce different dynamic behaviors within the same families of materials, we imposed significant diversification within the arrays, (i) by varying the interdigitated electrode spacing, which affects the linear to non-linear ratio of the voltage divider used for measurement and the diffusion of the analyte, (ii) by varying the form factor (which affects the number of “squares” in parallel) so to accommodate a larger number of sensing polymers with widely varying range of polymer resistivity. Gap sizes between electrodes ranged from 10-40 µm and sensor element areas ranged from 7200 µm² to 54400 µm² [9].

Currently, the system is composed of 16384 sensors of 25 different types. An extensive analysis is being carried out to assess that strategies we adopted effectively provide the artificial epithelium with the desired features. A semi-automatic system was developed to deliver well-controlled concentrations of pure compounds and binary
mixtures to 16384 sensors at the same time. Characterisation of redundancy and variability within the large array is here reported.

2.2. Results

The different polymers and the diversity of the characteristics of the transducer geometries contribute to generate a range of sensor response patterns. This is of fundamental importance in the realisation of a device mimicking natural olfaction. In order to verify the effectiveness of this assumption in the artificial epithelium, we analysed the response of all the sensors and grouped their responses depending on the polymer class they belong to.

Fig. 1, Fig. 2 and Fig. 3 depict boxplots representing the steady state ($\Delta R/R_0$) response of all the 16384 sensors to an input of 13734 ppm of ethanol, 5576 ppm of acetic acid and 19520 ppm of butanone.

Fig. 1 Boxplot of the steady state response of the sensors to a fixed concentration of ethanol.

Fig. 2. Boxplot of the steady state response of the sensors to a fixed concentration of acetic acid.

Fig. 3. Boxplot of the steady state response of the sensors to a fixed concentration of 2-butanone

Each separate box represents the statistical information about the sensor responses for each class of polymers. The numbers on the x-axes represent the polymer type, as coded in Table 1. The line included in each box depicts the mean of the sensor response for that particular type of polymer. Each box extends from the 25th to the 75th percentile of the sensor responses, while the whiskers indicate the most extreme sensor responses not considered as outliers. The boxplot representation of the response of the sensors suggests that the distribution of the sensor response of a polymer class is statistically different from the distribution of the sensor response of any other polymer class (as verified by performing an ANOVA test on different response classes). Similarly, the distribution of the response of a polymer group changes with different analytes.

This result demonstrates that even though the sensing materials used belong to three main polymeric families, the different doping, the addition of side chains and the realization of copolymers, modified their sensing characteristics, allowing us to produce an instrument with high variability and redundancy levels.
Table 1. List of the 25 polymers deposited onto the sensor substrate

| Class number | Polymer type      | Class number | Polymer type      |
|--------------|-------------------|--------------|-------------------|
| 1            | PANI/CB           | 14           | PANI/CSA/PVP      |
| 2            | PEDOT             | 15           | PANI/pTSA/PS      |
| 3            | PANI Long Chain   | 16           | PANI/pTSA         |
| 4            | PPy doped         | 17           | PANI/CSA/PEO      |
| 5            | PDDT/FeCl$_3$     | 18           | PANI/pTSA/PEO     |
| 6            | PANIPOL           | 19           | PANI/CSA/PS       |
| 7            | P3HT              | 20           | PANI/CSA/PVP      |
| 8            | PPy/CB            | 21           | PANI/CSA/PMMA     |
| 9            | PDDT/FeCl$_3$ modified | 22           | PANI/DBSA/PS     |
| 10           | PANI/CSA          | 23           | PANI/DBSA/PMMA    |
| 11           | P3DT/FeCl$_3$     | 24           | PANI/DBSA/PEO     |
| 12           | P3OT/FeCl$_3$     | 25           | Poly(3-methyl-4- hexylthiophene-2,5-diyl) |

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