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Electronic tongue for simultaneous determination of cyanide, thiocyanate and iodide



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ABSTRACT

Five cross selective electrodes comprising cyanide, thiocyanate, and iodide coated graphite electrodes CGEs were used as sensing parts of an electronic tongue for simultaneous determination of cyanide, thiocyanate, and iodide ions in the presence and the absence of chloride ion as a coexisting ion. The CGEs were constructed using poly(vinyl chloride) (PVC), o-nitrophenyl octyl ether (o-NPOE), cetyltrimethylammonium bromide (CTAB), and metal-porphyrin complexes as ionophores. The constructed electrodes were characterized to obtain their calibration curve, response time, and repeatability.

A programmed switching system combined with an array of the five potentiometric coated graphite electrodes (i.e., ion-selective or cross-selective electrodes) were connected directly to a pH/potentiometer and a computer to sequentially acquire the potential corresponding to water sample mixtures. The acquired potentials were recorded and saved on the computer and were used as input variables for an artificial neural network to simultaneously yield the concentrations of cyanide, thiocyanate and iodide ions in simple and complex mixtures. A feed-forward, back propagation network with a Levenburg–Maquart algorithm was employed to optimize the network parameters.

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1. Introduction

One of the most advantageous features of potentiometric sensors is their fast response that makes them convenient tools in the analysis of large number of samples. However, the cross selectivity of most of them sacrifice these characteristics, and make their use in complicated matrices difficult to some extent. As discussed in our previous work [1], application of some array of cross selective electrodes or the electrodes with low selectivity can compensate this failure.

Cyanide occurs naturally in many foods (e.g., cassava, flax, sorghum, bamboo shoots, and bitter almonds) and is

naturally generated by microorganisms (fungi, algae, and bacteria) [2]. The ISE methods for cyanide ion are subject to interferences from iodide, sulfide, bromide, thiosulfate, and thiocyanate, and they are also matrix dependent [3–5].

Thiocyanate and iodide are two of the most lipophilic anions which can penetrate into the PVC membrane, so that the membranes may show considerable potential responses toward them even if no ionophore is added into the membranes. These characteristics may be vital if the applied ionophores were not selective enough to the primary ion, so this makes the potentiometric sensors be cross-selective.

Determination of anions such as cyanide and thiocyanate in the presence of iodide may be necessary in clinical studies, as they can be found in biological samples. For example, thiocyanate contents may be found in the

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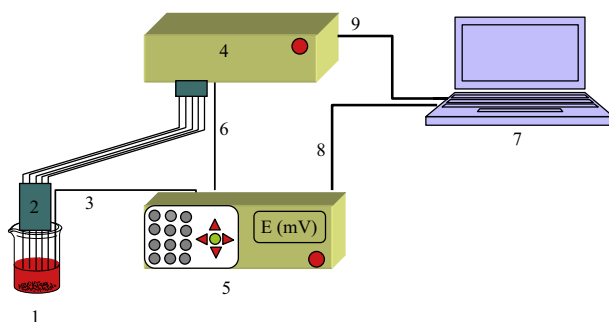


Fig. 1. Manifold of the electronic tongue, sample (1); arrays of sensors (2); reference electrode (3); switching system (4); pH meter (5); output of the switching system (input to the pH meter) (6); computer (7); serial connection of pH meter to computer (8); and serial connection of the switching system to computer (9).

presence of cyanide ion in biological fluids of smokers or other poisoned with cyanide ion [6,7]. On the other hand, iodide ion is also one of the nutrients needed for the endocrine gland, thyroid, to work properly [8]. Thus, the determination of iodide together with cyanide and thiocyanate in the biological sample is important. So, the simultaneous determination of cyanide, thiocyanate, and iodide ions was aimed in this work.

2. Experimental

2.1. Reagents and materials

Sodium or potassium salts of anions were purchased from Fluka or Merck, and were used without any further purification. High molecular weight PVC was prepared from Merck. All plasticizers were purchased from Fluka and used thoroughly. Metal-Schiff base and metal-porphyrin complexes were synthesized, and purified by using the procedure discussed in Refs. [9,10].

2.2. Electronic tongue configuration

A five ion-selective-electrode-array system was employed for obtaining potential of the samples containing the anionic species. The home-made instrument was used to acquire the potentials, sequentially (Fig. 1) [1]. In the current study, just five ports of the system were turned on. Thus in this case, the potential acquisition time was expected to be reduced, compared to the previous work, associated with the use of seven sensors. However, in the current study, it was tried to use cross selective electrodes which could contain information corresponding to all of the anionic species.

2.3. Electrode preparation

The membrane-coated graphite electrodes were prepared using the procedure discussed in the literature [1]. The optimized membrane compositions of the electrodes are summarized in Table 1. As it is seen, the electrodes nos. 1 and 4 are cyanide selective electrodes, electrodes nos. 2 and 3 are thiocyanate selective, and electrode no. 5 is an iodide selective electrode [9]. The electrodes were

conditioned in the solutions of ions of interest for at least 48 h.

2.4. Sample preparation

45 ternary sample mixtures of the anions with random concentrations of cyanide in the range of 1.0×10^{-2} M to 3.0×10^{-5} M, thiocyanate, and iodide in the range of 1.0×10^{-2} M to 1.0×10^{-5} M were prepared as calibration sample sets. Addition of some buffering solution was necessary to adjust pH of each of the samples in a proper pH, the hydrolysis of cyanide and thiocyanate being neglected. The optimized pH range for the two membrane coated graphite electrodes were 4.2–9.3 and 4.4–9.6 for thiocyanate selective electrodes. It was 9.5 for both cyanide selective electrodes and was about 2.0–9.5 for the iodide selective electrode. Consequently, all of the applied samples were buffered at a constant pH of 9.3, as all of the electrodes work conveniently at this pH, and the interference from OH^- ion in this region is also tolerable. Thus, 5 mL of borax buffer (0.1 M of pH 9.3) was added to each of sample solutions. After that, proper amounts of stock sodium cyanide, sodium thiocyanate, and sodium iodide solutions were added consecutively to the samples to obtain different random concentrations of the aforementioned anions. Finally, the samples were diluted to 25 mL; so, all of the samples had approximately a constant ionic strength.

In order to evaluate the analytical application of the array system, a set of 10 samples with different concentration of the aforementioned anions were made (Table 2). To check the reliability of the array configuration for predicting the anion concentrations, another 11 sample sets was also prepared with different random concentration of the anions in the presence of different concentration levels of chloride ion with a similar procedure discussed above (Table 3).

3. Results and discussion

3.1. Response time study

As mentioned earlier, sample matrices have such an intensive effect on the response of each electrode. So it was necessary to evaluate and average response time of

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