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Application of a Polypyrrole Sensor Array Integrated into a Smart Electronic Tongue for the Discrimination of Milk Adulterated with Sucrose

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Abstract. This work presents the use of a smart electronic tongue for discriminating adulterated milk samples with various concentrations of sucrose. The smart electronic tongue was integrated by a voltammetric sensor array from polypyrrole doped with various doping agents, a portable multi-potentiostat controlled by a smartphone with an Android application. Sucrose concentrations ranging from 1% to 20% were used to adulterate the tested samples. The sensor array was optimized to perform measurements on milk while maintaining good performance in terms of stability and signal quality. The sensor array was prepared by chronoamperometric electropolymerization of pyrrole with different doping agents, varying polymerization time (from 50 to 300 s), and concentration of monomer (from 0.10 to 0.40 M) and doping agent (from 0.05 to 0.30 M). The optimization process results demonstrated that the parameters polymerization time, monomer concentration, and doping agent concentration affect the stability of the signals in the sensors, allowing for the establishment of adequate conditions to guarantee maximum stability through an experimental design. Thus, values of 0.10 M for monomer concentration, 0.05 to 0.10 M for doping agent concentration, and 50 to 70 s for polymerization time were established. The measurements taken with the smart electronic tongue on the milk samples allowed a principal component analysis to classify the samples in the plane of the first two principal components. Principal components 1 and 2 registered a variance of 93.39% (78.68% and 14.71%, respectively), indicating a high degree of information registered by the sensor array. It could be concluded that the array of optimized polypyrrole sensors allows sufficient information to be recorded through measurements made with the smart electronic tongue to discriminate adulterated milk samples with different sucrose concentrations.

Keywords: Adulteration; Milk; Polypyrrole; Smart electronic tongue; Sucrose

1. Introduction

Milk is one of the most important natural foods for humans due to its high nutritional value. The United Nations Food and Agriculture Organization (FAO) considers it a complete and irreplaceable food for humans due to its nutritional composition (Guetouache *et al.,* 2014). Due to this, its consumption worldwide has increased significantly as it is considered a staple first need product in the daily diet, either as milk or any of its derivatives (Wang *et al.,* 2020). Being a product of high consumption, it is necessary to monitor its quality and

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safety in order to avoid serious health problems.

An alternative to control the quality of milk relies on four very substantial guidelines: constituents of milk, bacterial content, its appearance, and contains adulterants. The adulteration of milk is undoubtedly one of the most serious issues in the industry. One of the most common practices on producer farms is the addition of water and chemicals such as sucrose to increase profits (Yang *et al.,* 2020). The development of new markets as a consequence of free trade agreements between different milk-producing countries obligates governments and the dairy industry to provide excellent quality products; as a result, it is critical to identify foreign or rare substances in the standard composition of milk. The main tools used for the detection of adulterants are traditional chemical methods such as volumetric methods, infrared spectroscopy, atomic absorption spectroscopy, liquid chromatography-mass spectrometry, and nuclear magnetic resonance, among others (Song *et al.,* 2020; Azad and Ahmed, 2016; Dos-Santos *et al.,* 2012). All these techniques have been demonstrated to be effective in milk analysis. However, these techniques are typically performed with expensive equipment, highly qualified personnel, require sample pre-treatment, and lengthy analysis times. They cannot be performed in situ due to the equipment's large volume and weight.

The electronic tongue can be defined as an analytical instrument, which has proved its analytical capacity in various fields of the food industry and has been used to analyze wines, beers, coffee, tea, honey, and milk, which have grown in popularity in recent years. This type of device operates on systems comprised of three major components: a sensor array with cross selectivity, an electronic multichannel measurement device, and a data analysis unit utilizing multivariate or pattern recognition techniques (Vlasov *et al.,* 2005). The electronic tongue devices developed were based on different analytical principles, and various sensor arrays have been used (Arrieta *et al.,* 2020; Facure *et al.,* 2020; Zabadaj *et al.,* 2018; Lipkowitz *et al.,* 2018). However, electronic tongues based on electrochemical measurements have received widespread acceptance from the scientific community and the industry, particularly those based on potentiometric and voltammetric sensor arrays (Arrieta *et al.,* 2020; Oroian and Ropciuc, 2019; Lipkowitz *et al.,* 2018).

The main systems of potentiometric electronic tongues reported the use of selective ion or non-selective electrodes elaborated with various types of materials and have been used in many applications and have shown to be effective in the analysis of food products (Oroian and Ropciuc, 2019; Borges *et al.,* 2018; Witkowska and Kubota, 2016). However, these electronic tongues are limited to the detection of charged chemical species (of ionic nature); they are also highly sensitive to electronic noise and require a high cost in terms of electronic instrumentation and data acquisition.

On the other hand, voltammetric electronic tongues work with electrodes o sensors made of noble metals (platinum, gold, iridium, rhodium, and palladium) or modified electrodes (Arrieta *et al.,* 2020; Wei *et al.,* 2013; Alcañiz *et al.,* 2012). In contrast to potentiometric sensors, they are not limited to detecting charged species and possess desirable properties such as versatility, robustness, and simplicity (Hayat *et al.,* 2019; Khalil *et al.,* 2018). Nevertheless, systems based on metallic electrodes offer poorly resolved voltammetric signals with little information because the curves do not represent defined electrochemical processes by the concept (Winquist *et al.,* 2005). For this reason, several research groups have focused on improving and perfecting those voltammetric systems by applying various sensor modification strategies whose signals include better-defined electrochemical processes in order to obtain more information from the analyzed medium. Sensors have been modified using phthalocyanines, perylene derivatives, polypyrrole, and other substances (Arrieta *et al.,* 2020; Medina-Plaza *et al.,* 2015).

The miniaturization and modification of voltammetric sensors are common practices in analytical chemistry (Katseli *et al.,* 2020). However, very few works that focus on the modification and miniaturization of sensor arrays for electronic tongues in such a way that

they allow portability and extend their field of application to in situ analysis. This work has presented the application of an array of miniaturized and modified polypyrrole voltammetric sensors in analyzing milk adulterated by sucrose. The study of milk in situ is crucial in the dairy industry because it prevents harmful or adulterated milk from being carried to the plant. Currently, there is no technology of this type. Hence, this research investigates the possibility of implementing the technology to detect milk adulteration using a smart electronic tongue in our laboratory, which is equipped with a miniaturized sensor array coupled to a portable electronic device based on PSoC microchip technology and controlled by a Smartphone.

2. Methods

2.1. Reagents and Materials

The following reagents were used: pyrrole, sodium dodecylbenzene sulfonate, sodium sulfate, ammonium persulphate, potassium ferrocyanide, p-toluenesulfonic acid, anthraquinone-2,6-disulfonic acid disodium salt, lithium perchlorate, and sucrose. All the reagents used were of analytical quality and were purchased from Sigma-Aldrich (St. Louis, Missouri, USA). The solutions were prepared using ultrapure water of milli-Q quality (18 MΩ cm^{-1}). The samples analyzed were produced from the same sample of raw milk from Zebu cows (Bos taurus indicus). For this, an unadulterated sample (0%) and sucrose-adulterated samples (1%, 2%, 3%, 4%, 5%, 10%, and 20%) were taken for this purpose. There were five replicates for each sample, for a total of 40 samples (8 concentrations x 5 replicates each).

2.2. Smart Electronic Tongue Setup

The smart electronic tongue developed in our laboratory consisted of a polypyrrole voltammetric sensors array and a portable multi-potentiostat controlled by a smartphone. The sensor array was comprised of seven electrodes or voltammetric sensors of polypyrrole (PPy) doped with seven different doping agents (i.e., counterions): PPy/DBS (PPy doped with sodium dodecylbenzene sulfonate), PPy/SO4 (PPy doped with sodium sulfate), PPy/SF (PPy doped with ammonium persulphate), PPy/FCN (PPy doped with potassium ferrocyanide), PPy/TSA (PPy doped with p-toluenesulfonic acid), PPy/AQDS (PPy doped with anthraquinone-2,6-disulfonic acid disodium salt), and PPy/PC (PPy doped with lithium perchlorate). The preparation of the sensor array was conducted by chronoamperometric electropolymerization of pyrrole at 0.8 V, using an EG&G 2273 PAR potentiostat/galvanostat, controlled by the PowerSuite software. The PPy with each dopant was electrodeposited on graphite substrates arranged in a circular shape using an AC9C card of BVT Technologies.

The polymerization process was optimized by applying an experimental design in which the variables of greatest significance were evaluated to achieve the optimal parameters of polypyrrole synthesis in the elaboration of each sensor in the array. The experimental process corresponds to a 33-factorial design (three factors and three levels), totaling 27 treatments. The three factors were pyrrole concentration (0.10 M, 0.20 M, and 0.40 M), polymerization time (50 s, 100 s, and 300 s), and doping agent concentration (0.05 M, 0.10 M, and 0.30 M). The portable multi potentiostat was made on a FREESOC card using a PSoC 5LP microchip which was programmed with the PSoC creator software. This electronic device was designed to record the voltammetric signals of the seven sensors on the array simultaneously through seven measurement channels. In addition, a Bluetooth card was also included for data transmission to a Smartphone equipped with an Android application designed for device control and data recording. Details on electrochemical

polymerization techniques and electronic device development have previously been reported (Arrieta *et al.,* 2018; Arrieta and Fuentes, 2016; Arrieta *et al.,* 2015).

2.3. Measurement

Milk samples were measured at room temperature on 10 mL samples. The voltammetric signals were recorded at a sweep speed of 100 mV s^{-1} in a potential range of -1.0 V to 0.5 V with an initial potential of 0.0 V. Each sensor generated a voltammogram (i.e., voltammetric signal) of each sample, which was made up of 100 current data, allowing for 700 data points with the entire sensor array, each of which was a variable in the data matrix in each sample. When all samples were analyzed, a matrix of 28 000 data (700 variables x 40 samples) was constructed. The obtained matrix was analyzed using the principal components analysis method to determine the discrimination ability of the smart electronic tongue on this type of sample.

3. Results and Discussion

Once the sensor array was elaborated, it was integrated into the device to test its proper functioning and the correct registration of the voltammetric signals through the seven channels. Figure 1 depicts a smart electronic tongue. It can be seen the three components that make it up as well as an expansion of the sensor array where the reference electrode of Ag/AgCl of the array (RE), the counter electrode (CE), and the array of seven sensors (S1…S7) can be observed.

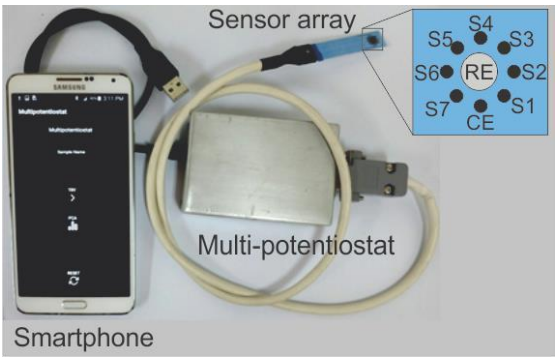

Figure 1 Image of the smart electronic tongue formed by the miniaturized sensor array, the portable electronic device, and a Smartphone with an Android application for device control

To guarantee the proper functioning of the sensors, their response to a raw milk sample was initially explored; all the sensors were synthesized using a polymerization time of 70 s, with monomer and doping agent concentrations of 0.20 M and 0.10 M, respectively. Voltammetric signals were obtained without noise and with well-defined peaks during the measurements. However, the sensors showed signals with some loss of intensity, which could affect the measurements made with the smart electronic tongue. As an example, Figure 2 demonstrates the response of the S7 sensor (PPy/AQDS). It can be seen that the signal in the first recorded cycle had a current intensity of 508.8 uA. After 50 consecutive cycles, the signal reached a current intensity of 470.4 uA, representing a loss of intensity of 7.54%. For the other sensors, the intensity losses recorded were 5.23%, 8.25%, 0.71%, 4.84%, 1.12%, 3.40% for PPy/SO4, PPy/DBS, PPy/SF, PPy/FCN, PPy/PC and PPy/TSA, respectively. As previously described in other projects that evaluated the response of polypyrrole sensors in various types of products, the loss of intensity in the signals could be caused by the exit of the doping agent from the polypyrrole matrix, which can be

modulated by factors such as the concentration of monomer and doping agent used in the synthesis solution and the polymerization time (Arrieta *et al.,* 2020; Foroughi *et al.,* 2009).

Although the current losses do not exceed 9%, which represents a low loss due to the sensors, in this case, no more than 50 voltammetric cycles should be registered, and process optimization of the sensor elaboration process was carried out to generate a more stable and consistent signal over time. Therefore, monomer and doping agent concentration, as well as polymerization time, were used to carry out the optimization process by applying an experimental design in which the loss of intensity in the signals, represented as a percentage of stability, was used as the output variable.

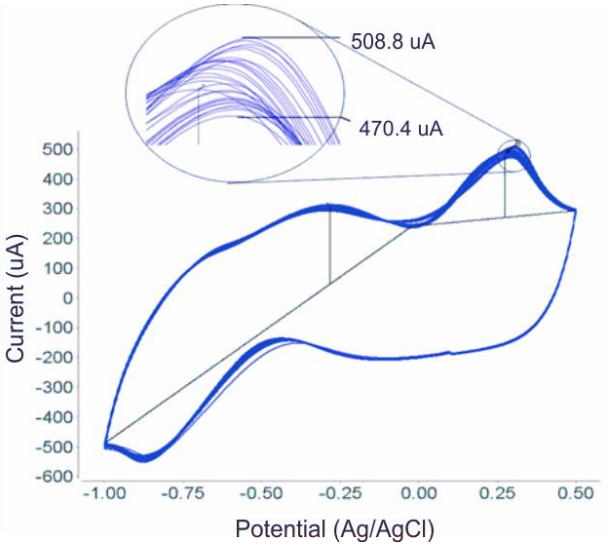

Figure 2 Voltammetric signal of the PPy/AQDS sensor registered in a raw milk sample

The results of the optimization experiment design revealed that the analyzed factors influence the signals' stability and the fact that there are interactions between them. Table 1 displays the results of the variance analysis performed with the PPy/TSA sensor. The statistical significance of the factors and their interactions can be appreciated, specifically polymerization time, the interaction between polymerization time and concentration of the doping agent, and the interaction between concentrations of monomer and doping agent used in the electrodeposition process. In this case, it is possible to conclude that these factors significantly impact the sensor signal's stability. These effects had P values less than 0.05, indicating that they significantly differed from zero at a 95 % confidence level. The most significant factor is the interaction between the concentration of pyrrole and the concentration of the doping agent. It was very important to determine an optimal value for these factors because they can significantly interfere with the sensor array's performance when analyzing milk samples.

The interactions and behavior of the factors against stability were observed in the response surface graphs obtained from the optimization study, as shown in Figure 3. Response surface graphs and level curves were utilized to analyze the effects of polymerization time, pyrrole concentration, and doping agent concentration on signal stability (i.e., loss of signal intensity), which was used as the response variable. This was done to obtain the optimal values of the levels for each factor used in elaborating each sensor in the array.

It was possible to determine that the dopant concentration presents optimum stability at intermediate values close to 0.10 M. The concentration of pyrrole for extreme values tends to increase the sensors' instability. In this sense, the instability increases because of

the rise in time and is offset by a high or low dopant concentration. As for the time, the trend exhibits greater instability using prolonged reaction times. It can also be seen that the lowest areas of the response surface have the most suitable values to carry out the elaboration of the sensors, which correspond to polymerization times of less than 100 s, pyrrole concentration between 0.10 and 0.20 M, and a dopant concentration between 0.05 and 0.10 M.

Table 1 Variance analysis of the factors; polymerization time (t), the concentration of monomer [Py], and concentration of doping agent [DA] obtained in the optimization process of the PPy/TSA sensor

Source of variation	Sum of Squares	GI	Mean Squares	F	P-Value
A:Time (t)	169.341	1	169.341	6.24	**0.0230**
B:[Py]	3.25976	1	3.25976	0.12	0.7331
C:[DA]	5.95125	1	5.95125	0.22	0.6454
AA	2.87042	1	2.87042	0.11	0.7489
AB	52.0833	1	52.0833	1.92	0.1837
AC	172.521	1	172.521	6.36	**0.0219**
BB	4.335	1	4.335	0.16	0.6943
BC	431.28	1	431.28	15.90	**0.0010**
CC	73.2902	1	73.2902	2.70	0.1186
Error	461.064	17	27.1214		
Total (corr.)	1376.0	26			

Based on the optimization results, the polymerization time values and concentrations of monomer and pyrrole to be used in the elaboration of each sensor were established. Table 2 illustrates the conditions used while also showing the new values for the percentage of intensity loss obtained by optimizing the processing parameters. It can be observed that the stability improved remarkably, and the intensity losses did not exceed 3% in any of the sensors, with very low values of 1% were obtained in the case of PPy/SO4 and PPy/AQDS.

After the sensor array was optimized, the measurements were carried out on the milk samples to evaluate the ability of the smart electronic tongue to classify samples adulterated with sucrose. The data matrix for the principal component analysis was created from the signal registers. The correlation circle generated by the principal components analysis is depicted in Figure 4. Figure 4 shows that the data recorded in the signals generated by the sensor array provide a large amount of information because most of the points in the correlation circle are distributed at values close to 1 and -1 of the quadrants; that is, the circle has a radius close to 1, indicating that the sensor array provides a large amount of helpful information for sample discrimination. In addition, it can be seen that the sensors do not provide redundant information; this is evidenced by the fact that the areas of overlap of points are scarce in the circle, inferring that all of the sensors provide important information about the samples analyzed.

The variance or information captured in the first two components was 93.39%, the first principal component with 78.68%, and the second principal component with 14.71%. Figure 5 displays a score graph demonstrating the discrimination of the samples of milk adulterated with sucrose. It can be clearly seen that the samples with the same amount or concentration of adulteration (i.e., sucrose) are clearly grouped together, forming clusters corresponding to the various concentrations of adulterant studied. Furthermore, the clusters are well differentiated from one another, establishing clear discrimination of the samples analyzed.

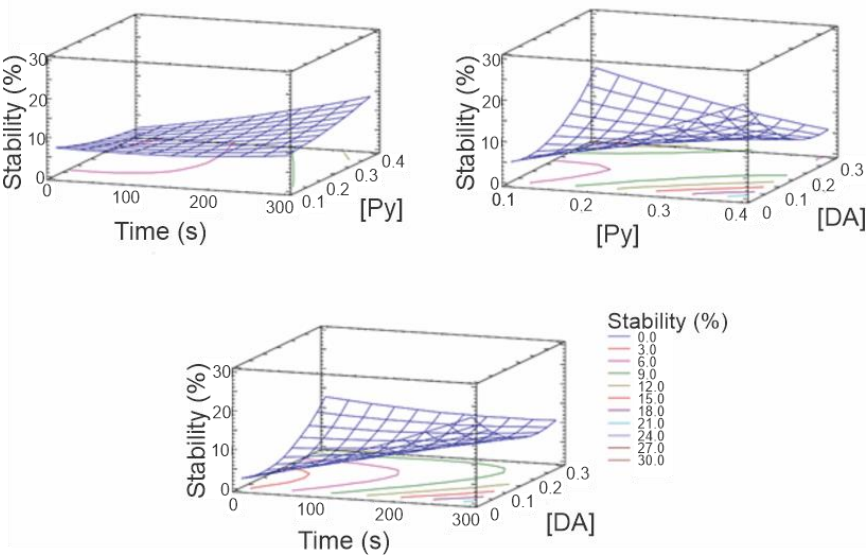

Figure 3 Response surface of the most significant processing factors for the PPy/TSA sensor

Table 2 Parameters used in the process of elaboration of the sensor array obtained from the optimization process

Sensor	Acronym	Monomer/Doping agent concentration [M]	Polymerization time (s)	Loss of intensity (%)
S1	PPy/SO4	0.10/0.05	55	0.39
S2	PPy/DBS	0.10/0.10	50	2.50
S3	PPy/SF	0.10/0.05	65	3.00
S4	PPy/FCN	0.10/0.10	60	1.98
S5	PPy/PC	0.10/0.10	60	2.15
S6	PPy/TSA	0.10/0.10	70	2.56
S7	PPy/ AQDS	0.10/0.05	60	0.91

The unadulterated samples (0%) and the adulterated with 1% are separated in the negative quadrant of the two components (quadrant -: -). The sample adulterated with 2% is found in the negative quadrant of the first component and positive in the second component (-: + quadrant), while the groups of adulterated samples with 3%, 4%, 5%, and 10% are found in the positive quadrants of both components (quadrant +: +). The sample with the largest amount of adulterant (20%) is separated into the positive quadrant of the first component and the negative quadrant of the second component (quadrant +: -).

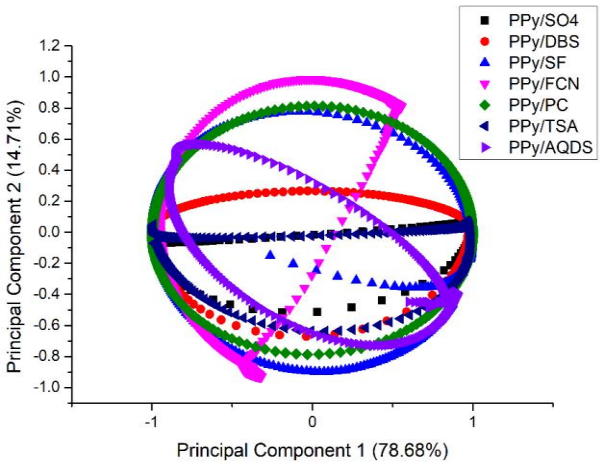

Figure 4 Correlation circle of the variables generated by the sensor array registered in the principal component analysis

Although samples with 4% and 5% sucrose concentrations touch each other, they do not overlap so that they can be distinguished easily. Thus, each sample group is well discriminated, and the smart electronic tongue could classify each sample based on its adulterant content.

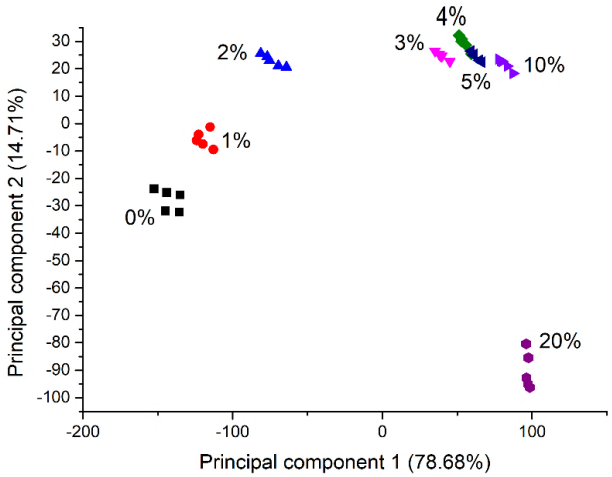

Figure 5 Correlation circle of the variables generated by the sensor array registered in the principal component analysis

4. Conclusions

By using a smart electronic tongue integrated with a polypyrrole sensor array, a portable measurement device, and a smartphone made it possible to carry out measurements on adulterated milk samples, obtaining well-defined signals. The elaboration parameters of the sensors, such as polymerization time, monomer concentration, and doping agent concentration, affect the stability of their voltammetric signals against milk samples. Thereby, an optimization process had to be carried out by applying an experimental design that allowed obtaining well-defined signals with high stability. The optimal value of monomer concentration was 0.10 M, the polymerization time was between 50 and 70 s, and the doping agent concentration was between 0.05 and 0.10 M. The measurements on the sucrose-adulterated samples allowed to register signals that contained enough information to be able to differentiate them. Principal components 1 and 2 recorded a total of 93.39% variance, with 78.68% in principal component 1 and 14.71% in principal component 2. Thus, the result of the principal component analysis demonstrated the discrimination capacity of the smart electronic tongue on sucrose-adulterated samples.

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