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Research Article

Detection of Adulteration in Argan Oil by Using an Electronic Nose and a Voltammetric Electronic Tongue

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Adulteration detection of argan oil is one of the main aspects of its quality control. Following recent fraud scandals, it is mandatory to ensure product quality and customer protection. The aim of this study is to detect the percentages of adulteration of argan oil with sunflower oil by using the combination of a voltammetric e-tongue and an e-nose based on metal oxide semiconductor sensors and pattern recognition techniques. Data analysis is performed by three pattern recognition methods: principal component analysis (PCA), discriminant factor analysis (DFA), and support vector machines (SVMs). Excellent results were obtained in the differentiation between unadulterated and adulterated argan oil with sunflower one. To the best of our knowledge, this is the first attempt to demonstrate whether the combined e-nose and e-tongue technologies could be successfully applied to the detection of adulteration of argan oil.

1. Introduction

Argan oil is a vegetable oil that has recently conquered the world. Argan oil, which is prepared by press extraction of argan kernels [1], is a typical Moroccan product. Indeed, the argan tree is exclusively endemic in Morocco and is not grown anywhere else [2]. The argan forest is an 800,000 ha large area covering the fertile Souss valley region, the foothills of the Anti-Atlas mountains, and the coastal region between Essaouira and Agadir. Argan oil is prepared from the fruits of argan trees (Argania spinosa (L.) Skeels) following a multistep process [3]. This oil exists as comestible and/or cosmetic. When the kernels contained in the argan fruits are slightly roasted prior to grinding, comestible argan oil is obtained. However, the unroasted kernels are saved to prepare oil dedicated to cosmetology. For centuries, the production of virgin argan oil has played an invaluable economic role in Morocco, and today the total annual production reaches approximately 4,000 tons per year [4]. Argan oil is a relatively new international product that nowadays is exported only by Morocco. It can be assumed that exports will increase in a near future due to the unique properties of the product [5]. The price of argan oil in Europe is approximately 100 € per liter, being considered a luxury food. Due to its price, the possibility of illegal practices such as dilution with cheaper oils is frequently encountered.

Adulteration is defined as the process by which the quality or the nature of a given substance is reduced through the addition of a foreign or an inferior substance and the removal of a vital element [6]. The adulteration of high-quality oil with cheap vegetable ones is a frequent problem from regulatory agencies, oil suppliers, and consumers. Several methods have been proposed for the detection of the adulteration of high-quality oils such as, high performance liquid chromatography (HPLC) [7], solid phase microextraction gas chromatography-mass spectrometry (SPME-GC-MS), time

of flight mass spectrometry (MALDI-TOF-MS) [8], Raman spectroscopy [9], near infrared spectroscopy [10], fluorescence [11], and nuclear magnetic resonance [12]. Recently, argan oil adulteration has been also the subject of some research works; inductively coupled plasma optical emission spectrometry (ICP-OES) have been used [6, 13]. However, most of these techniques are usually time-consuming and costly for routine use in food industry; hence there is a large demand for rapid, cheap, and effective techniques for food quality control and especially for food detection adulteration.

For this purpose, techniques based on e-noses and e-tongues, which approximately mimic human nose and taste bud, respectively, have been used to characterize edible oils and deal with adulteration problems in vegetable ones [14–23]. Indeed, the e-noses/tongues have the ability to discriminate different vegetable oils and detect adulteration in some cases. However, there is no report on the application of the e-noses/tongues targeting the fraud detection in virgin argan oil. To our knowledge, this is the first time that an e-nose and an e-tongue have been applied to quantify the percentages of adulteration in virgin argan oil. This paper examines the ability of an e-nose and a ve-tongue for the recognition of adulteration percentages defined as the amount of sunflower oil added to pure argan oil. The data that are produced by the electronic systems are considered as an overall fingerprint, which can be interpreted with the use of powerful pattern recognition methods such: principal component analysis (PCA), discriminant factor analysis (DFA), and support vector machines (SVMs).

2. Materials and Methods

2.1. Sample Preparation and Measurements. The quality of the handmade argan oils used in this study is guaranteed since they were obtained directly from the producers. Argan oil was used in its rough state, without any preliminary processing. For the adulteration study (performed in our lab), adulterated oil samples were prepared by mixing argan oil (A.O) (comestible and cosmetic) with the sunflower oil (S.O), leading finally to six different samples at every concentration level ((A.O : S.O) (100 : 0), (90 : 10), (70 : 30), (50 : 50), (30 : 70), and (0 : 100)). Six measurements were made in succession for each type of oil mixture for e-nose and e-tongue analysis. Since six percentages of adulteration in virgin argan oil were analysed, the data set has included 36 samples. Measurements of the oil headspace and taste were carried out, respectively, by means of an e-nose and a ve-tongue which will be described in Sections 2.2 and 2.3.

For the e-nose measurements, each sample of argan oil was introduced into airtight glass vials and kept under dark conditions just before use. The samples (10 mL) were heated at 32 ± 1°C, inside a controlled thermostat-sampling chamber for a headspace generation time of 10 min. This yielded a homogeneous headspace and no transfer of argan oil headspace had occurred yet. The e-nose sampling system consisted of a dynamic headspace sampling. In this way, the volatile compounds were directly transferred by the carrier gas into the sensor chamber. The vial had two small holes in their covers to allow the headspace to be analyzed within the e-nose equipment. For each set of oil that was analyzed, a new airtight glass vial was used.

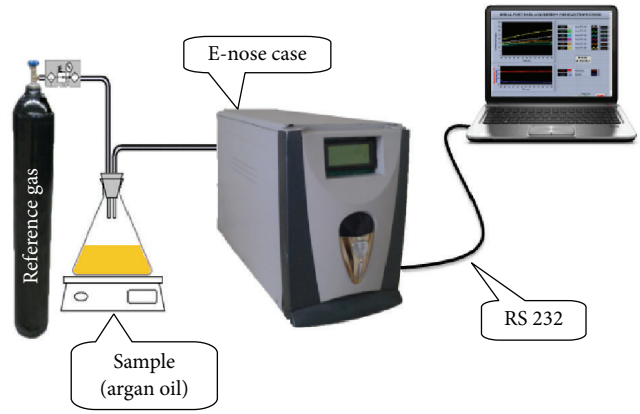

Figure 1: E-nose setup for adulteration detection of Moroccan argan oil.

Electrochemical measurements were thermostatically controlled using a water bath of ~32°C. The cyclic voltammograms were recorded in a range of varying potential from −600 to 800 mV and with a scanning rate of 20 mV s^{-1}. An electrochemical cleaning step was also performed to prevent the accumulative effect of impurities on electrode surface. After each reading, the electrodes spend 2 minutes in a solution of 50 mM KOH and 25% H_2O_2 before rinsing with distilled water [24].

The stability of the proposed systems and the repeatability of the measurements were not measured explicitly. However, no significant response drift was observed during this experimental period for the raw value of the TGS sensors conductance measured in the presence of the reference gas and for the voltammetric electrodes in KCl solution after each electrochemical cleaning step. A P value <0.05 was the significant threshold.

2.2. E-Nose Setup. A MOS gas electronic nose system based on a 5-sensor array was used [25]. The experimental system is mainly composed of three parts: the sampling system, the sensor chamber, and the data acquisition system. An overall view of the system is shown in Figure 1. The sensor array comprised five different tin-dioxide gas sensors: TGS 8xx (with xx = 15, 22, 24, 25, and 42) obtained from Figaro Engineering, Inc. (Osaka, Japan), a temperature sensor (LM335Z), and a relative humidity sensor (HIH4000-01) from National Semiconductor. Data acquisition system was accomplished using a PIC16F877 microcontroller and Lab VIEW© software (National Instruments Inc., Austin, Texas, USA) program running on a laptop PC.

2.3. VE-Tongue Setup. The e-tongue consisted of seven working electrodes, an Ag/AgCl (3 M saturated KCl, diameter 2 mm) reference electrode, and a platinum auxiliary electrode (length 5 mm, diameter 2 mm), and these were arranged in a standard three-electrode configuration. The seven working electrodes were: platinum (Pt), gold (Au), glassy carbon (GC), silver (Ag) (all purity 99.9%, length 5 mm, and diameter

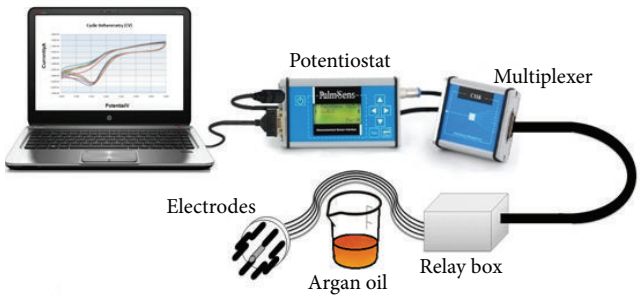

Figure 2: VE-tongue setup for the adulteration detection of Moroccan argan oil.

2 mm) from CH Instruments, Texas, USA; nickel (Ni), palladium (Pd), and copper (Cu) (all purity 99.9%, length 6 mm, and diameter 1.6 mm); these electrodes were made by BAS Inc., Tokyo, Japan. All the seven working electrodes were embedded in composite material placed around the auxiliary electrode, and only the edges of the working electrodes and the auxiliary electrodes were exposed. The wires from the electrodes were connected via a relay box to a portable potentiostat PalmSens (PalmSens BV, The Netherlands). The cyclic voltammetry is applied as the measurement principle in this study. In cyclic voltammetry, the potential of working electrode is varied linearly with time, while the reference electrode maintains a constant potential. The potential is applied between the reference electrode and the working electrode and the current is measured between the working electrode and the counter electrode. In the present work, the output current is considered for analysis. The voltage pulses are applied with the help of a potentiostat PalmSens and the entire setup is controlled by a personal computer. The voltammetric electronic tongue setup with three-electrode configuration is shown in Figure 2.

2.4. Feature Extraction and Data Preprocessing. The purpose of feature extraction is to attain a low-dimensional mapping that preserves most of the information in its original feature vector. To analyze the response of the e-nose data, two parameters were extracted from each sensor conductance transient: the steady-state conductance (G_S), calculated from the average value of the conductance during the last minute of the measurement and the dynamic slope of the conductance (dG/dt), calculated between 2 and 7 minutes of the exposure time to the argan oil samples. Since there were 5 gas sensors within the array, each measurement was described by 10 features. For the ve-tongue, it is well-known that voltammetric signals contain hundreds of measures and they usually have overlapping regions with nonstationary characteristics. In order to fully exploit the information that was obtained from each voltammogram, three representative features from the cycle voltammogram of each sensor of the array were extracted. The complete list of these features is $\Delta I = I_{max} - I_{min}$, the current change calculated as the difference between maximum and minimum values of the current, S_{ox}, the maximum slope of the current curve in the oxidation shape, and S_{rd}, the maximum slope of the current curve in the reduction shape. As there were 7 working electrodes within the array, each voltammetric measurement was described by 21 variables. The choice of these e-nose and ve-tongue features was based on our previous works [26]. The e-nose and ve-tongue data have been normalised in order to remove any concentration effects in the sample. A mean-centering preprocessing technique was then applied to the datasets.

2.5. Data Analysis. To show the response of the e-nose and e-tongue, both chemical and electrochemical responses were used to form databases which were subjected to unsupervised and supervised multivariate data analysis methods such as PCA, DFA, and SVMs.

PCA is a very well-known unsupervised method often employed with several kinds of gas and taste sensor arrays [27]. The main objective of PCA consists of expressing information contained in a dataset by a lower number of variables called principal components. These principal components are linear combinations of the original response vectors. The principal components are chosen to contain the maximum data variance and to be orthogonal. Hence, PCA allows the reduction of multidimensional data to a lower dimensional approximation, while simplifying the interpretation of the data by the first two or three principal components (PC1, PC2, and PC3) in two or three dimensions and preserving most of the variance in the data.

DFA is probably the most frequently used supervised pattern recognition method and the best-studied one [28]. DFA is based on the determination of discriminant functions, which maximize the ratio of between-class variance and minimize the ratio of within-class variance. As in PCA, this technique is a factorial method. In fact, using this method, data are separated in k a priori defined classes. The objective sought using DFA is to investigate if the variables are sufficient or not to allow a good a posteriori classification of data in their a priori groups.

In the last decade, a new classification technique called SVMs has been proposed in the broad learning. SVM is a supervised learning technique for the building of classifiers. SVMs have been shown to be a powerful learning method [29] and their use by the e-nose and e-tongue communities has gained importance in the last years. SVMs were originally designed for binary classification. Currently there are two types of approaches for multiclass SVM. One consists of constructing and combining several binary classifiers "one-against-one or one-against-all methods", while in the second approach, all data are directly considered in a single optimization formulation. The latter constructs during the learning phase a hyperplane that separates the different classes, which enables the inferring of a class amongst the k previously learnt when a new input is presented during the prediction phase. Full algorithm details can be found elsewhere [30].

3. Results and Discussion

3.1. Adulteration Detection of Argan Oil Using the E-Nose

3.1.1. E-Nose Responses. The volatile compounds generated from the argan oil headspace were pumped at a flow of

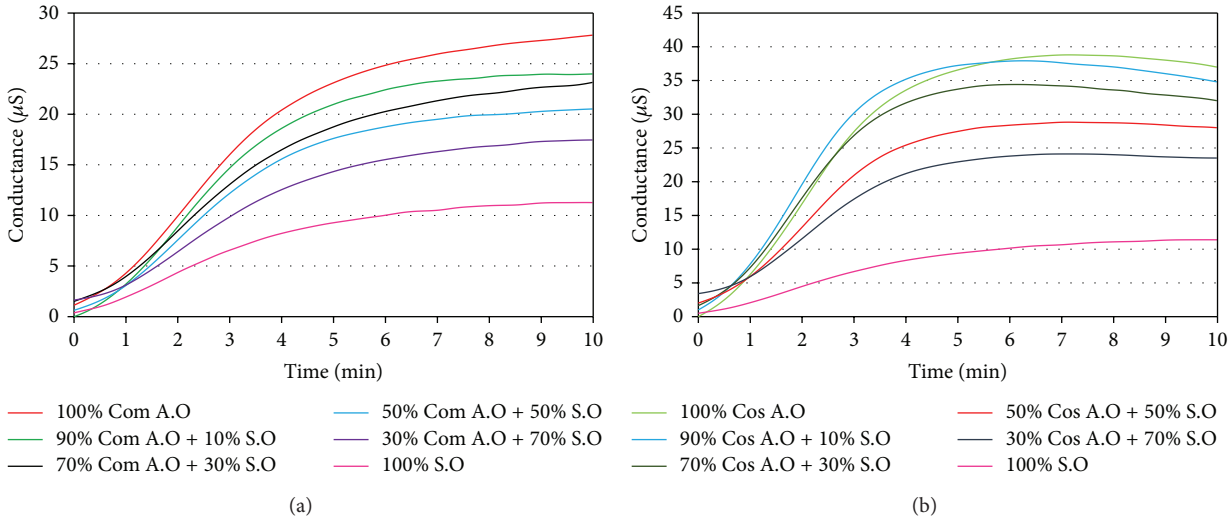

Figure 3: Time conductance evolution of the TGS 842 sensor for (a) comestible and (b) cosmetic argan oil.

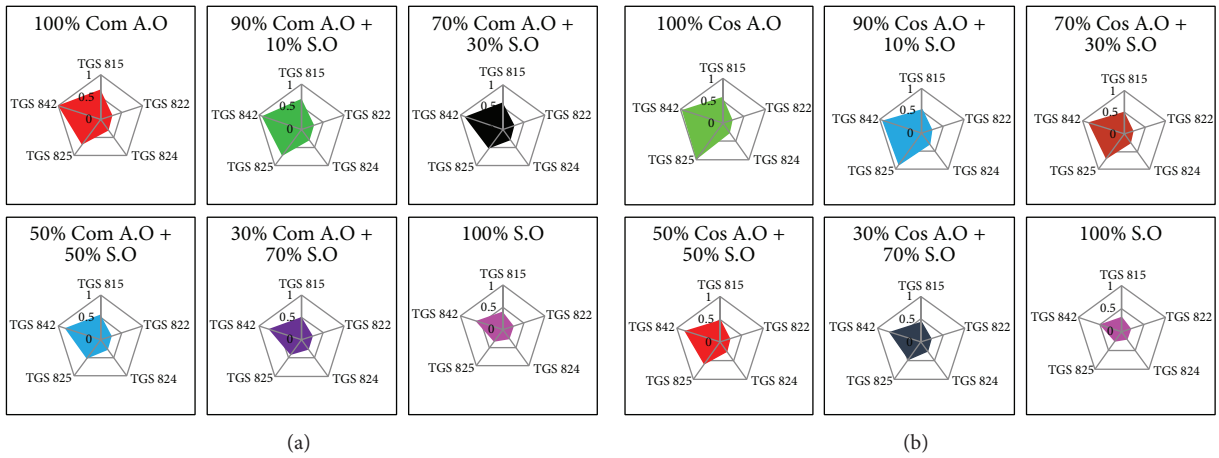

Figure 4: Radar plots of the steady-state conductance of the sensor array for (a) comestible and (b) cosmetic argan oil.

1000 sccm through the measurement chamber containing the array of the 5-gas sensors. Upon injecting the sample, data were acquired every 2 s over 10 min. Figure 3(a) shows typical recordings of TGS 842 gas sensor towards comestible argan oil (Com.A.O), sunflower oil (S.O), and comestible argan oil adulterated with different proportions of sunflower oil (10 to 70%). Idem, Figure 3(b) shows typical recordings of the same sensor towards cosmetic argan oil (Cos.A.O), sunflower oil (S.O), and cosmetic argan oil adulterated with different proportions of sunflower oil (10 to 70%). It is observed that the signals reach a plateau after 10 min of measurement. Moreover, it can be observed in Figure 3 that the sensor response of TGS 842 has been decreased drastically depending on the percentage of the sunflower oil added. A flushing time of 5 min was enough to desorb the volatiles from the sensors and enable the signals to return to the baseline.

Radar-like plots with unitary radius were used to observe whether pattern differences (i.e., fingerprints) were developed among the different percentages of sunflower oil added to the pure argan oil. Figure 4 shows a representative case for both Com.A.O (Figure 4(a)) and Cos.A.O (Figure 4(b)). To construct these plots, the values of TGS steady-state conductance were divided by the value corresponding to TGS 842 which showed the maximum signal. This helps to easily visualize the differences amongst typical response patterns. As can be seen, a clear pattern variation can be observed for unadulterated argan oil and those adulterated with sunflower.

3.1.2. PCA and DFA Results. In order to test the capability of the developed e-nose to recognize the degree of adulteration of argan oil, we have analyzed their data by means of PCA and DFA. The PCA procedure was performed using MATLAB 7.0.1 software (MathWorks Inc., Natick, Massachusetts, USA). A mean centring preprocessing technique was applied to the response matrix. Figure 5 shows the three-dimensional score plot which accounts for 98.81% in the case of comestible argan oil and their adulteration (Figure 5(a)) versus 98.09% in the case of the cosmetic argan oil and their adulteration (Figure 5(b)) of the total variance, respectively.

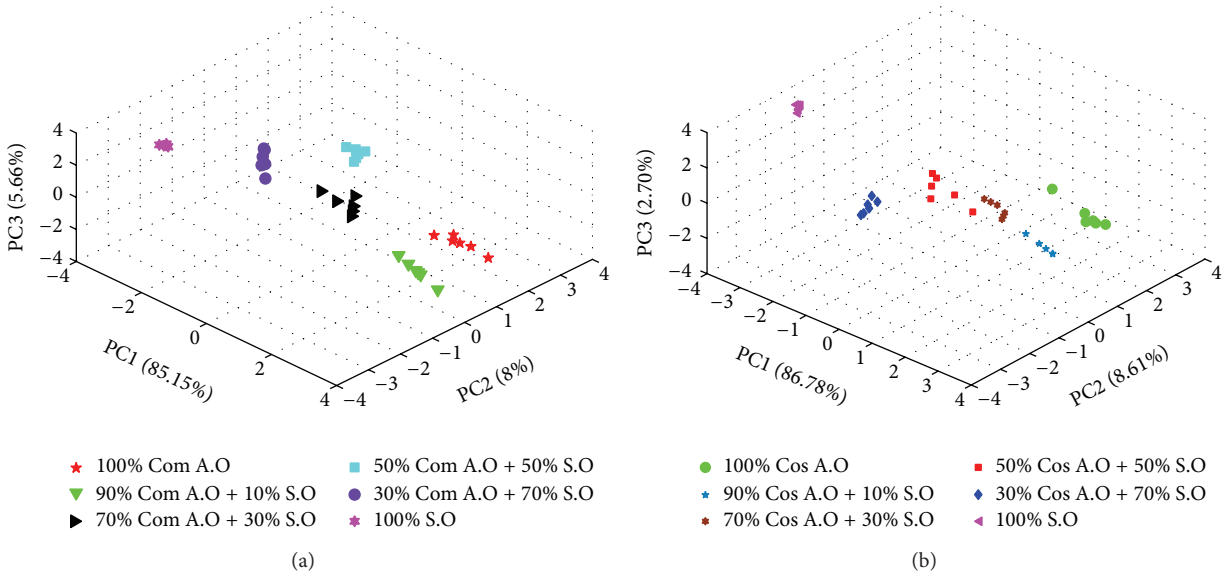

Figure 5: Scores plot of a PCA performed on (a) comestible and (b) cosmetic argan oil measurements gathered using the e-nose.

In both PCA plots, six distinct groups, corresponding to the comestible/cosmetic argan oil, sunflower oil, and argan oil adulterated with different proportions of sunflower oil (10 to 70%) can be easily identified.

The DFA is amongst the best classification methods, since this procedure maximized the variance between categories and minimized the variance within categories in order to optimize the resolution. The model was built using all the available samples as training set, in order to check the classification capability of DFA leave-one-out cross-validation approach. The procedure of DFA was performed by means of SPSS software, and its results are shown in Figure 6(a) for the Com.A.O and in Figure 6(b) for the Cos.A.O. It can be seen that each oil group is correctly discriminated. It can be seen that each oil mixture is correctly classified. The two first discriminant functions show perfect classification of the six groups.

3.1.3. Prediction of Adulteration by SVM. SVMs and the one-against-one classification method were applied to develop the classifier model. A second-order polynomial kernel function was used to project the training data to a space that maximizes the margin hyperplane. The optimal regularization parameter of the SVM (C) was set to infinity, which implies that no classification errors were tolerated for the set of response patterns employed for training the SVMs. Due to the relatively small number of measurements available, leave-one-out cross-validation method was implemented to better estimate the true success rate that could be reached with the SVMs. This assumes that, with the given n measurements, the model was trained n times using $n - 1$ training vectors. The vector left out during the training phase was then used for the test. The performance of the given model was estimated as the average performance over n tests. Table 1(a) shows the confusion matrix of the SVM classifier for comestible argan oil and Table 1(b) for the cosmetic one. Results indicated that 91.67% and 83.34% of success rate were achieved in the recognition of the comestible and cosmetic argan oil, respectively.

In order to further enhance the discrimination of argan oil samples and also their adulteration levels, a voltammetric e-tongue has been investigated.

3.2. Adulteration Detection of Argan Oil Using the Voltammetric E-Tongue

3.2.1. VE-Tongue Voltammograms. The cyclic voltammetry CV was used to study the qualitative information generated by the electrochemical process. The CV measurements were carried out from −0.6 V to +0.8 V with a scan rate of 0.02 Vs^{-1}. Under these conditions, the voltammetric sensors showed a variety of anodic and cathodic peaks. Figures 7(a) and 7(b) report the CV voltammograms of the Pt sensor immersed in comestible and cosmetic argan oil, sunflower oil, and argan oil adulterated with different proportion of sunflower oil (10 to 70%), respectively. As can be observed, different response profiles were obtained depending on the degree of the adulteration of comestible and cosmetic argan oil (not only in the voltammogram shape, but also in the obtained currents). The presence of clear and poor redox peaks (one cathodic peak at around −0.2 V) reveals changes in the voltammograms and indicates that different redox species can be present in the argan oil matrix. The same behavior was also observed for the other sensors with different anodic and cathodic peak positions and intensities.

Radar plots of the normalized data for the seven voltammetric sensors, expressed as the slope of oxidation S_{ox}, are shown in Figure 8. For unadulterated Com.A.O and their adulterated Com.A.O (Figure 8(a)) as well as for unadulterated Cos.A.O and their adulterated Cos.A.O (Figure 8(b)) with S.O, a characteristic sensing fingerprint is generated.

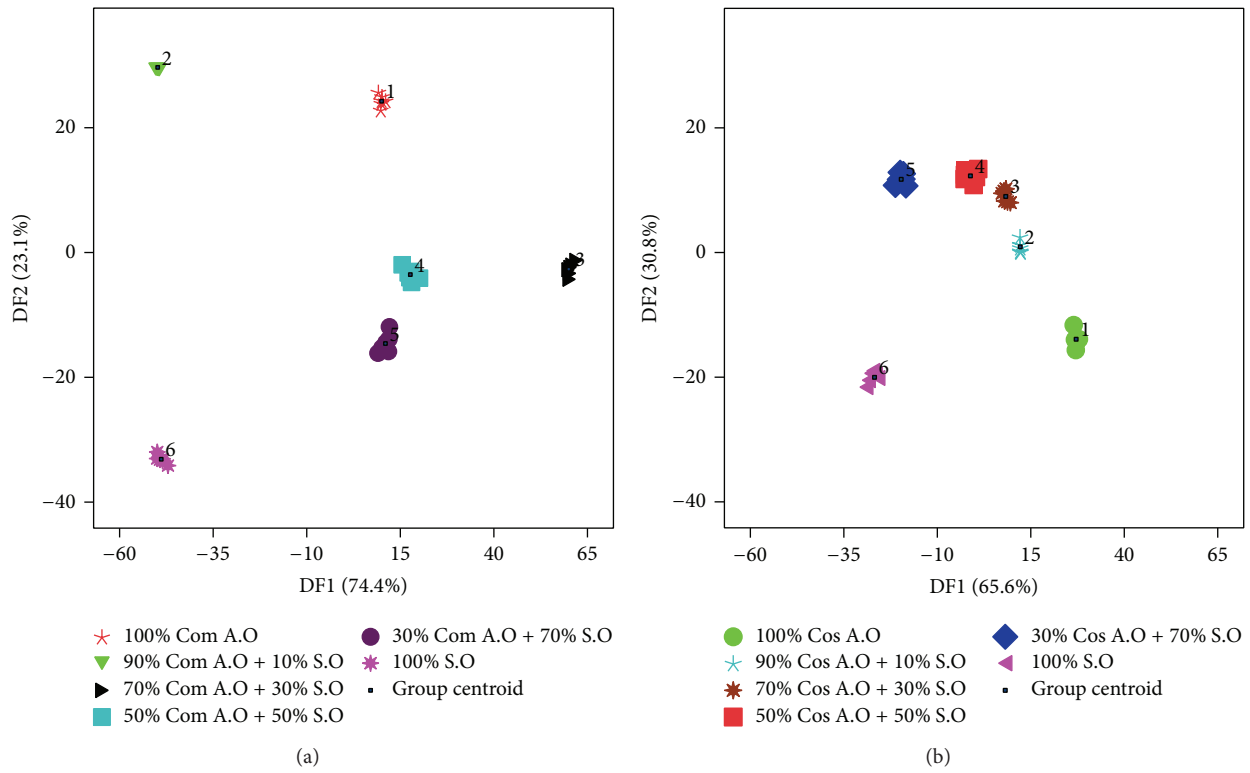

Figure 6: Discriminant factor analysis (DFA) plot for (a) comestible and (b) cosmetic argan oil using the e-nose.

Table 1: SVM classification results using the e-nose.

(a)

Actual	Predicted					
	100% Com A.O	90% Com A.O + 10% S.O	70% Com A.O + 30% S.O	50% Com A.O + 50% S.O	30% Com A.O + 70% S.O	100% S.O
100% Com A.O	5		1			
90% Com A.O + 10% S.O		6				
70% Com A.O + 30% S.O	2		4			
50% Com A.O + 50% S.O				6		
30% Com A.O + 70% S.O					6	
100% S.O						6

(b)

Actual	Predicted					
	100% Cos A.O	90% Cos A.O + 10% S.O	70% Cos A.O + 30% S.O	50% Cos A.O + 50% S.O	30% Cos A.O + 70% S.O	100% S.O
100% Cos A.O	4	1	1			
90% Cos A.O + 10% S.O		5	1			
70% Cos A.O + 30% S.O			5	1		
50% Cos A.O + 50% S.O			1	4	1	
30% Cos A.O + 70% S.O					6	
100% S.O						6

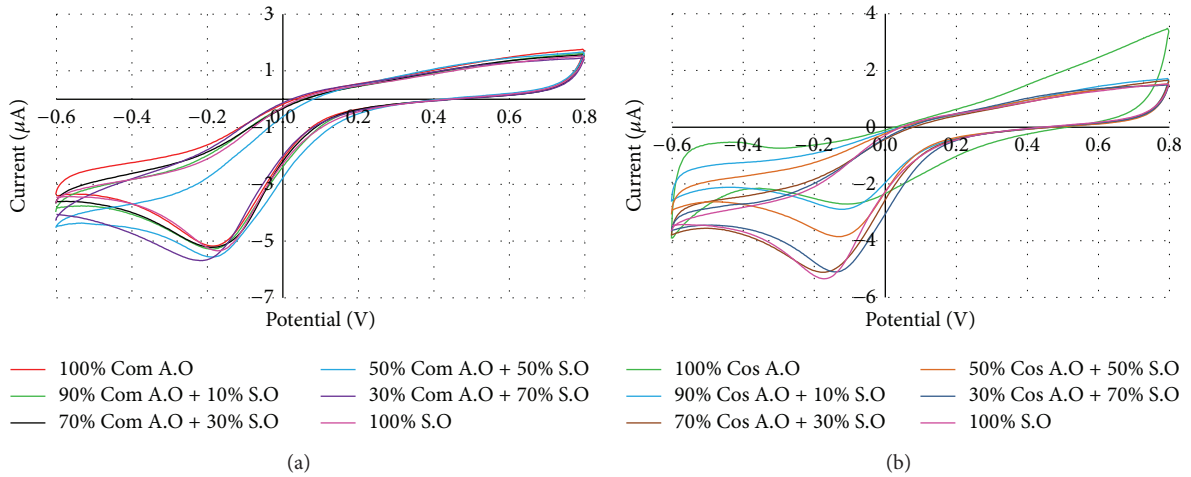

FIGURE 7: Voltammetric responses of Pt sensor immersed in samples of different percentage of sunflower oil added to (a) comestible and (b) cosmetic argan oil.

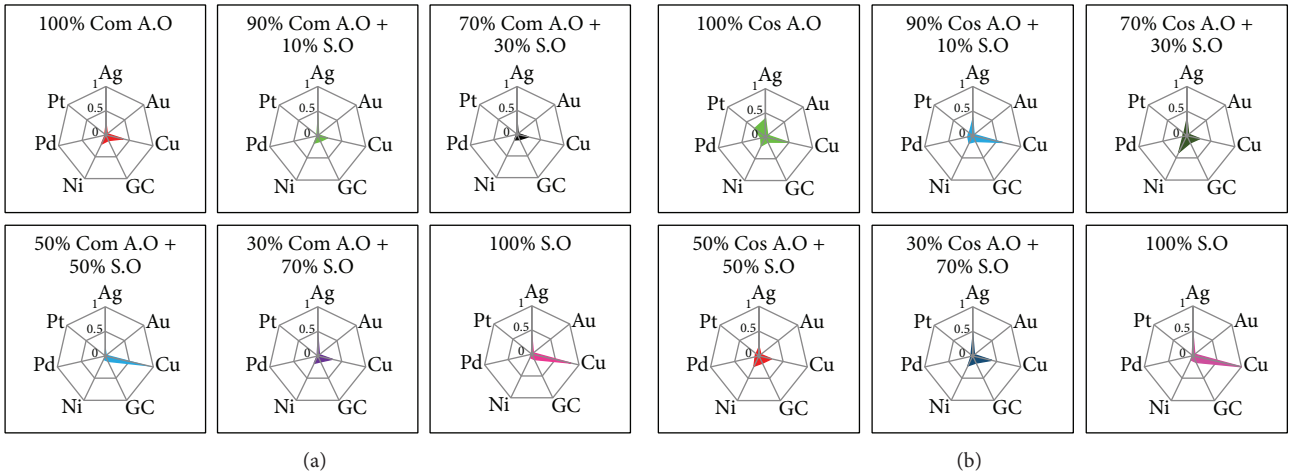

FIGURE 8: Radar plots of the responses of the array of voltammetric sensors (expressed as the oxidation slope S_{ox}) for (a) comestible and (b) cosmetic argan oil.

In fact, the radar plots show at a glance a clear pattern variation between the different oils.

3.2.2. PCA and DFA Results. PCA and DFA have been applied to discriminate and classify the argan oil adulterated with different proportions of sunflower oil. First, the discrimination capacity of the ve-tongue was evaluated by means of PCA. Figure 9 shows the scores plot of the three first principal components calculated from the three aforementioned extracted features (ΔI, S_{ox}, and S_{red}). The first three principal components explained 74.04% of the information (PC1 = 33.60%; PC2 = 26.24%; and PC3 = 14.2%) in the database corresponding to comestible argan oil (Figure 9(a)), and 78.27% of the information (PC1 = 37.80%; PC2 = 24.87%; and PC3 = 15.92%) in the database corresponding to cosmetic argan oil (Figure 9(b)). Both PCA score plots indicated that the voltammetric electronic tongue was able to discriminate among pure argan oil, adulterated ones, and sunflower oil. Therefore, a perfect recognition of all the adulteration levels was reached.

Applying DFA, a good separation between oils samples was obtained. Figure 10 shows how the first two DFA functions discriminate among clusters. DFA model was cross-validated using leave-one-out approach. An accuracy of 97.23% success rate in the recognition of the pure comestible argan oil, adulterated ones, and sunflower oil was achieved. On the other hand, 94.45% of the correct classification has been reached by DFA classifier for the pure cosmetic argan oil, adulterated ones, and sunflower oil.

3.2.3. Prediction of Adulteration by SVM. SVMs and the one-against-one classification method were applied to develop the classifier model. Like for the PCA and DFA, 21 response features from the voltammetric sensor array were used as inputs to the SVMs. Second-order polynomial kernel functions were used to project the training data to a space that maximized

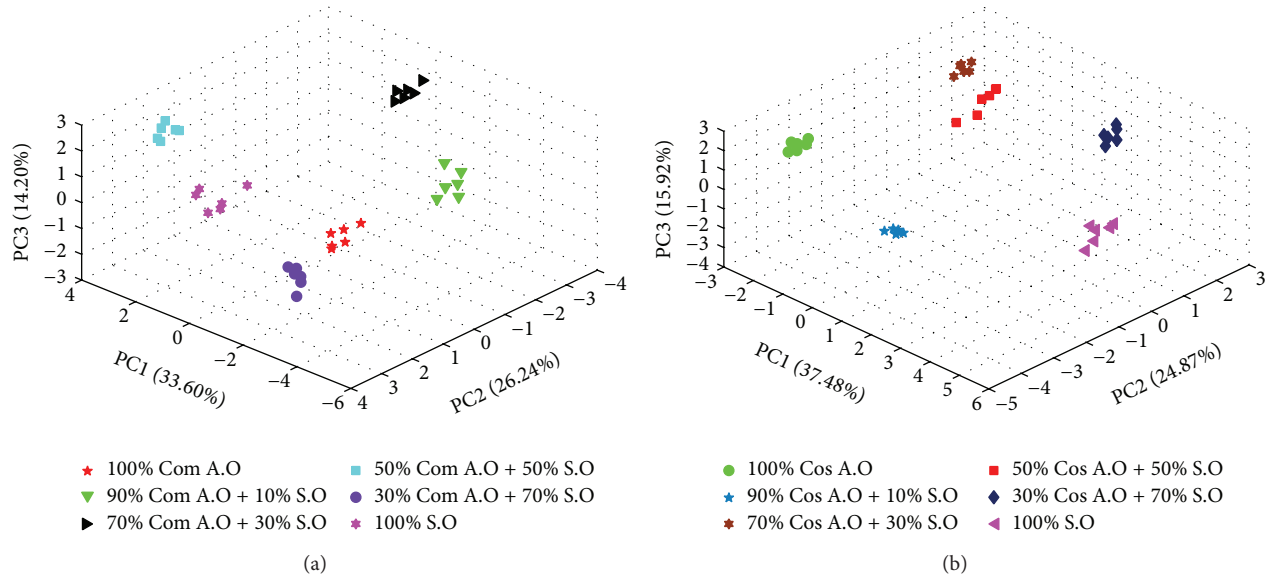

FIGURE 9: Scores plot of a PCA performed on (a) comestible and (b) cosmetic argan oil measurements gathered using the voltammetric e-tongue.

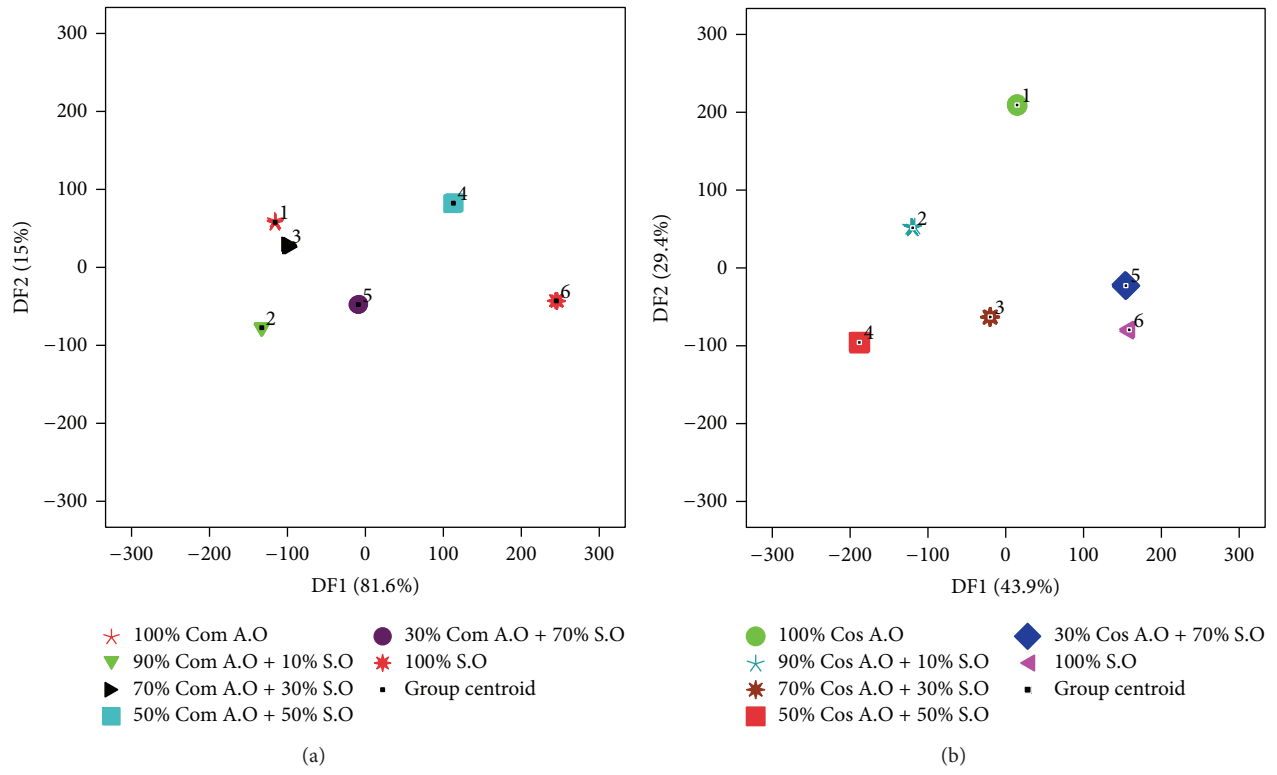

FIGURE 10: Discriminant factor analysis (DFA) plot for (a) comestible and (b) cosmetic argan oil using the ve-tongue.

the margin hyperplane. The optimal regularization parameter of the SVM was set to C = inf. The performance of the SVM model was evaluated using a leave-one-out cross-validation method. At first, a mean centring preprocessing technique was applied to the two datasets. For each dataset, six binary classification models were built (i.e., six percentages of argan oil groups). Then, the output of SVMs classifier was calculated as the most voted category. The SVM reached a 100% success rate in the recognition of both comestible and cosmetic argan oil. Table 2(a) shows the confusion matrix of the SVM classifier for the comestible argan oil and Table 2(b) shows the confusion matrix of the cosmetic argan oil.

Table 2: SVM classification results using the ve-tongue.

(a)

Actual	Predicted					
	100% Com A.O	90% Com A.O + 10% S.O	70% Com A.O + 30% S.O	50% Com A.O + 50% S.O	30% Com A.O + 70% S.O	100% S.O
100% Com A.O	6					
90% Com A.O + 10% S.O		6				
70% Com A.O + 30% S.O			6			
50% Com A.O + 50% S.O				6		
30% Com A.O + 70% S.O					6	
100% S.O						6

(b)

Actual	Predicted					
	100% Cos A.O	90% Cos A.O + 10% S.O	70% Cos A.O + 30% S.O	50% Cos A.O + 50% S.O	30% Cos A.O + 70% S.O	100% S.O
100% Cos A.O	6					
90% Cos A.O + 10% S.O		6				
70% Cos A.O + 30% S.O			6			
50% Cos A.O + 50% S.O				6		
30% Cos A.O + 70% S.O					6	
100% S.O						6

4. Conclusion

For the first time, an e-nose system based on 5-TGS sensors and an e-tongue formed by 7-voltammetric electrodes were employed to detect argan oil adulteration. Initially, PCA and DFA results dealing with the e-nose data showed acceptable results for the differentiation between pure and adulterated argan oils. Furthermore, SVM based classifier and the one-against-one voting strategy was implemented, which achieved acceptable success rates; 91.67% for comestible argan oil and 83.34% for cosmetic argan oil. The strength of the voltammetric e-tongue has been confirmed by its ability to correctly discriminate argan oil, sunflower oil, and argan oil adulterated with different proportions of sunflower oil (10 to 70%). Perfect recognition was achieved by PCA, DFA, and SVMs (the SVM model leading to a 100% success rate). In the light of these results, the proposed measurement systems represent excellent tools for the detection of the adulteration in argan oils and allow a perfect identification of the cheaper oil percentages added to pure ones.

Conflict of Interests

We declare that we do not have any commercial or associative interest that represents a conflict of interests in connection with the work submitted.

Acknowledgments

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