Fuzzy ARTMAP for the Adulterated Honey Discrimination with Voltammetric Electronic Tongue

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Abstract: In this paper, in order to improve recognition rate of the Jing Hua honey by adding different proportions of glucose, the pattern recognition methods of Radial Basis Function (RBF), Fuzzy k-nearest neighbor algorithm (FKNN) and Fuzzy Adaptive Resonance Theory MAP (Fuzzy ARTMAP) were used to classify the different honey adulterated proportion. The result shows that the recognition effect by using Fuzzy ARTMAP to discriminate the different concentration honey adulteration is better than RBF neural network and FKNN, and the recognition rate improved from 83.33 % to 94.40 %, and the discriminated speed of using Fuzzy ARTMAP is faster than RBF and FKNN. Fuzzy ARTMAP is a kind of recognition method which allows for fast and reliable. In addition, the prediction of honey adulterated proportion can be realized by The Partial Least Square (PLS).

Keywords: Fuzzy ARTMAP, Voltammetric electronic tongue, Honey Adulteration, PLS, RBF, FKNN.

1. Introduction

Honey is a natural substance that bees produce by gathering and combining flower nectar, secretions or honeydew with their own secretions, after transformation, dehydration, and storage within nest spleen to be matured. Its nutrition and medical value has attracted thousands of consumers. In recent years, due to the businessman in the pursuit of profit, honey adulteration often happened, which the main method of adulteration is mixed the honey with sucrose, glucose, starch, etc. [1]. Some manufacturers even use glucose, glucose syrup, essence replace raw materials to manufacture fake honey to profiteering. It is not only damage the interests of the consumer, but also affect the import and export trade of a country. Therefore, establishing effective rapidly honey adulteration detection method is an important part of the study of food safety and honey production.

There are many ways to research the quality of honey. However, traditional physical and chemical methods [2-3] are trivial, complex operation and low precision, high performance Liquid Chromatography (HPLC) [4], which has a lower sensitivity, and is greatly influenced by temperature. Capillary electrophoresis method, isotope mass spectrometry [5] and the near infrared spectroscopy [6] are able to rapid test but the price is more expensive, and it is difficult to spread in the small and medium-sized enterprises. Therefore, this topic is proposed to use electronic tongue for rapid non-destructive testing, which can save time and money [7-8]. Electronic tongue (ET) is a new intelligent instrument which is composed of sensor array, electrochemical workstation and personal computer. As a new intelligent instrument, electronic tongue has been applied to many sides, such as tea beverages [9-10],
alcohol [11-12], milk, meat, microbes and the environmental monitoring [13-17]. In addition, electronic tongue also has been applied to research of honey quality, with L. Gil-Sánchez and E. Garcia-Breijo [18] have done the recognition research into honey from different plants using the methods of Principal Component Analysis and Artificial Neural Network Algorithm by the potentiometric electronic tongue system. Z. B. Wei and J. Wang [19] have classified monofloral honey from different plants using more frequency and large amplitude pulse voltammetry by voltammetric electronic tongue system, using three pattern recognition methods: Principal Component Analysis, Discriminant Function Analysis and Cluster Analysis.

The neural network [20] is an information solution system to simulate the brain structure and function, which has already been widely used in the system modeling, pattern recognition, image processing and control. When the feed-forward neural network is used, we need to choose the optimal network size and parameters according to some heuristic methods. Besides, the network after learning usually is static. On the contrary, Adaptive Resonance Theory (ART) neural network family overcome the so-called plasticity, stability reorientation problem, and can learn new information in a changing environment. Fuzzy ARTMAP neural network is composed of two ART modules; the two modules are link through a Map Field. Because of this Map Field, the network can be very stable for arbitrary data input mode of real-time supervision and learning, and finally achieve the result which is indeed [21]. The neural network structure is clear, and classification accuracy is high, it has been widely used in the study of land cover classification and remote sensing image classification, and has achieved good classification effect.

In this paper, PCA is used to prove the efficiency of the data of the adulterated honey. RBF neural network, Fuzzy K-Nearest Neighbor and Fuzzy ARTMAP neural network are used to distinguish the different proportion of the adulterated honey. Partial Least Square (PLS) is used to predict the different proportion of the adulterated honey.

2. Experimental

2.1. Voltammetric Electronic Tongue System

The voltammetric electronic tongue system [22] consists of the sensor array, electrochemical workstation (CHI660C), personal computer (PC), which is shown in Fig. 1. The sensor array was replaced by standard three electrode system that the working electrode is gold electrode, the reference electrode is the saturated calomel electrode and the auxiliary electrode is platinum electrode. Electrochemical workstation (CHI660C) was manufactured by Shanghai Chenhua Instrument Limited Company. White, red and green three electrodes lead of electrochemical workstation respectively connected reference electrode, auxiliary electrode and working electrode. At some later time, the other side attached to the computer and established a connection. Then we insert the three electrodes in the solution to be detected, and choose the appropriate experimental method and set the appropriate parameters in the computer software. Electrochemical workstation can be used to generate a series of electrical pulses and pick up the response signals, on the other hand, it collected the response signal and stored response signal that experiment generated by the computer.

![Fig. 1. Voltammetric electronic tongue system.](image)

2.2. The Principle of Voltammetric Electronic Tongue

The principle of voltammetric electronic tongue is built on the basis of the electrochemical reaction, and different results are received by the different electrochemical detection methods. The voltammetric behavior of the system was studied by differential pulse voltammetry and linear sweep voltammetry, cyclic voltammetry, square wave voltammetry. The voltampere measuring principle is to apply a voltage between the working electrode and the reference electrode, to measure the resulting current between the working electrodes and the auxiliary electrode, to extract different information under the different current waveforms. Different concentrations of honey adulteration are measured by differential pulse voltammetry, the parameters setting is shown in Table 1.

<table>
<thead>
<tr>
<th>High Voltage (V)</th>
<th>Low Voltage (V)</th>
<th>Increment Voltage (V)</th>
<th>Pulse Width (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.2</td>
<td>0</td>
<td>0.008</td>
<td>0.01</td>
</tr>
<tr>
<td>Sample Width (s)</td>
<td>Pulse Period (s)</td>
<td>Quiet Time (s)</td>
<td>Sensitivity (A/V)</td>
</tr>
<tr>
<td>0.005</td>
<td>0.02</td>
<td>2</td>
<td>1e-6</td>
</tr>
</tbody>
</table>
2.3. Data Acquisition

Preparation of honey adulteration: Selecting Jinghua honey and Glucose. Honey is purchased from the supermarket in Jilin of China. The proportions of Jinghua honey are: 0 %, 10 %, 20 %, 30 %, 50 % and 70 %. The experiment is conducted at room temperature. Specific steps:

**Step 1:** Washing some beakers with deionized water.

**Step 2:** The samples were pretreated. Making sure the sample’s capacity is 300 ml by using deionized water, and Stirring for 30 seconds straight.

**Step 3:** The three-electrode regards soaking with alcohol for 5 minutes, followed by rinsing with deionized water and drying with filter paper. The electrode mount is fixed with the electrochemical workstation.

**Step 4:** Open electrochemical workstation switch. Technology and related parameters are required to set a good experiment and then the three electrodes are inserted into the liquid and to start to measure.

**Step 5:** Each sample is measured for 30 times. Repeat the steps, after all the test, 180 groups of data were received.

The maximum current value, the second maximum, the minimum current value and the second minimum were selected as the analysis signal of the electronic tongue system. The representative voltammograms corresponding to each kind of honey adulteration are shown in Fig. 2 (a-f).

(a) Differential pulse voltammetric curve (the proportion of Honey adulteration is 0 %).

(b) Differential pulse voltammetric curve (the proportion of Honey adulteration is 10 %).

(c) Differential pulse voltammetric curve (the proportion of Honey adulteration is 20 %).

(d) Differential pulse voltammetric curve (the proportion of Honey adulteration is 30 %).

(e) Differential pulse voltammetric curve (the proportion of Honey adulteration is 50 %).

(f) Differential pulse voltammetric curve (the proportion of Honey adulteration is 70 %).

Fig. 2. Differential pulse voltammetric curves for six different concentration of honey adulteration.
3. Data Processing and Analysis

3.1. Principal Component Analysis

Principal component analysis algorithm [23-24] is one of the widely used methods of electronic tongue system analysis of data processing. The calculation process of this algorithm is to construct a series of linear combination of the original variables, so as to achieve the maximum of the variance. It is also a method of reflecting the information of the original variables under the principle of uncorrelated each other, and reducing data redundancy as much as possible. The first couple of components of this group of new data contain most of the sample space information, so we can choose several components instead of the whole information. The status of principal components in the sample was performed by applying PCA to the standardized data and plotting the graph of their scores.

Classification results of different concentrations of honey adulteration using PCA is shown in Fig. 3. This different information is well distinguishable on the two first principal components, respectively. PC1, which explains 81.80 % of the variance contained in the original data, PC2, which explains 15.11 % of the variance contained in the original data. The cumulative contribution rate of the first two principal components is 96.91 %. The results show that PCA can distinguish different concentration honey adulteration.

![Fig. 3. The PCA score plot for honey adulteration.](image)

3.2. RBF Neural Network

Radial Basis Function Neural Network (RBF) [25-26] is a kind of efficient feed-forward neural network, which is simple in structure, easier for training and widely used. According to the test data nature of the adulterated honey, the structure of RBF neural network is 4*custom*5, training error is 0.0001, training time is 0.2 s. The number of training data is 126, and the number of testing data is 54. Classification results of different concentrations of adulterated honey using RBF neural network is shown in Fig. 4. The predict value is represented by "*" and the true value is represented by "o". If the two symbols overlap, unknown samples are correct, in contrast to identify the wrong samples. The result of using RBF neural network indicated that 9 of 54 samples of different concentration honey adulteration were misidentified, and the correct rate reaches 83.33 %.

![Fig. 4. Identification results of different concentration honey adulteration based on RBF.](image)

3.3. Fuzzy K-nearest Neighbor Algorithm

In 1985, Fuzzy K-nearest neighbor (FKNN) [27] was first proposed by Keller, Gray and Givens as a classification algorithm. The fundamental idea of FKNN is that k objects nearest to the testing data were found from the training data, then the classification of the testing data were determined by each neighbor category labels. Giving the different weighted coefficient of k objects nearest, the coefficient of the largest class label was calculated by using fuzzy decision as the category of the testing data.

The nearest neighbor number K and fuzzy parameter M of FKNN are two of the most important parameters. In Fig. 5, when K is 3, M is 2, the number of training samples is 126, only 6 samples out of 54 testing ones are identified correctly and the correct rate is 88.89 %.

![Fig. 5. Identification results of different concentration honey adulteration based on FKNN.](image)
3.4. **Fuzzy Adaptive Resonance Theory Map**

The concept of fuzzy ARTMAP neural network [28] is to introduce Fuzzy set theory to ARTMAP system, which can easily realize the mapping between input data and output data. The new minimum and maximum learning rule is achieved by matching tracking process, which are the minimum prediction error and the maximum coding compression rate. Fuzzy ARTMAP neural network consists of two ART modules, namely ARTa and ARTb, which are joined over a Map field. For any sequence of input mode can study for the stability of the real-time learning, and finally get the desired results. The structure of Fuzzy ARTMAP Neural network is shown in Fig. 6.

![Fig. 6. The structure of Fuzzy ARTMAP Neural network.](image)

Fuzzy ARTMAP steps are as follows:
1) Initializing parameter of the Fuzzy ARTMAP;
2) The ARTa module accepts input data by using the complement and encoding;
3) The winning neuron is determined by calculating the largest selection function;
4) Adjust the vigilance parameter, if it meets the requirements, matching output vector of the map filed;
5) If the match succeeds, network resonance occurs, and adjusts to the weight values of winning neuron, then continue to Step 7;
6) If the two don't match, the matching tracking is executed, and then continues to Step 3;
7) If there is no learning model, then return to Step 2, whereas learning algorithm is done.

The vigilance parameter of Fuzzy ARTMAP neural network is 0.85, choice parameter is 0.000001, and learning rate is 1. Fig. 7 presents the Fuzzy ARTMAP neural network result. When the number of training samples is 126, only 3 samples out of 54 testing ones are identified correctly and the correct rate is 94.40 %.

![Fig. 7. Identification results of different concentration honey adulteration based on Fuzzy ARTMAP.](image)

3.5. **Results and Discussion**

The recognition of honey adulterated proportion using three methods of pattern recognition is shown in Table 2. The correct of discrimination with RBF is 83.33 %, the correct of discrimination with FKNN is 88.89 %, but the correct of discrimination with Fuzzy ARTMAP is up to 94.40 %, in addition, the speed of discrimination with Fuzzy ARTMAP is better than RBF and FKNN.

<table>
<thead>
<tr>
<th>Pattern recognition method</th>
<th>Recognition rate</th>
<th>Time (s/10)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RBF</td>
<td>83.33 %</td>
<td>1.61</td>
</tr>
<tr>
<td>FKNN</td>
<td>88.89 %</td>
<td>2.54</td>
</tr>
<tr>
<td>Fuzzy ARTMAP</td>
<td>94.40 %</td>
<td>0.63</td>
</tr>
</tbody>
</table>

4. **Honey Adulteration Proportion Forecast**

Partial Least Square (PLS) is a new kind of method of multivariate statistical data analysis. It originally is a dependent relationship variable synthesis two or more independent variable algorithm. Partial least square can forecast data are divided into several regions, then based on the characteristic vector describing each area of the same area, vector interaction orthogonal, different areas, vector will be based on prior channel and the model related [29].

In this paper, the prediction of honey adulterated proportion can be realized by PLS. The correlation coefficient of PLS is 0.8442, the mean square error is 0.2414, and the number of principal components is 3. Fig. 8 shows the prediction of honey adulteration and the error is 4.5 %.

5. **Conclusions**

The voltammetric electronic tongue system based on three-electrode system was used the adulterated honey discrimination. Feature extraction of the data was acquired by differential pulse voltammetry, and the PCA was used to deal with these data. Then Fuzzy ARTMAP, RBF and FKNN were used to classify the testing data.
The result shows that the recognition effect by using Fuzzy ARTMAP to discriminate the different concentration honey adulteration is better than RBF neural network and FKNN, and the recognition rate improved from 83.33% to 94.40%, and the discriminated speed of using Fuzzy ARTMAP is faster than RBF and FKNN. The prediction of honey adulterated proportion can be realized by PLS. It can be concluded that Fuzzy ARTMAP is a kind of fast, effective and reliable recognition method. It will continue to expand the other food quality detection and identification field in the future.

Acknowledgments

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References


[29]. B. Li, Study on dam safety monitoring statistic models based on partial least squares regression, *Xi’an University of Technology*, 2007.

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