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A PROPRIETARILY DEVELOPED BIONIC OLFACTORY SYSTEM USED FOR RAPID DETECTION OF DETERIORATED REFRIGERATED-STORED APPLES

Hui Tian,* Wenshen Jia,*,**,***,**** Jie Ma,* Jihua Wang,**,***,**** and Jianxiong Hao*****

Abstract

To reduce huge economic losses caused by apple deterioration to growers and dealers, a high-sensitivity, low-cost portable electronic nose detection system used for rapid detection and early-warning of apple deterioration was designed. The system comprises an electrochemical sensor array, data collection and transmission module and human–machine interaction software. Firstly, data collected by the electronic nose was pre-processed with smoothing filtering procedures, then a non-destructive detection model for refrigerated-stored apples was constructed on the basis of bionic olfactory technology in combination with bionics, principal component analysis (PCA), algorithms of kernel principal component analysis (KPCA), backpropagation neural network (BPNN) and support vector machine (SVM). The experiment results show that seven-point smoothing filtering can produce better outcomes, and models on the basis of backpropagation (BP), SVW, PCA+SVM and KPCA+SVM all reach a correct recognition rate of more than 90%, with correct recognition rates of PCA+SVM being higher than those of BP, SVM and KPCA+SVM. This research provides significant reference value for research focusing on electronic nose applied in non-destructive rapid detection and early-warning of apples.

Key Words

Electronic nose, sensor, principal component analysis, bakepropagation neural network

* College of Automation, Beijing Information Science and Technology University, Beijing, China; e-mail: yth13145@126.com; {jiawenshen; mjbeijing}@163.com

** Department of Beijing Research Center for Agricultural Standards and Testing, Beijing Academy of Agriculture and Forestry Sciences, Beijing, China; e-mail: wangjh@brcast.org.cn

*** Department of Risk Assessment Lab for Agro-products (Beijing), Ministry of China, Beijing, China

**** Key Laboratory of Urban Agriculture (North China), Ministry of Agriculture and Rural Affairs, Beijing, P.R. China

***** College of Bioscience and Engineering, Hebei University of Science and Technology, Hebei Shijiazhuang, China; e-mail: cauhjx@163.com

Corresponding author: Wenshen Jia

Recommended by Dr. Dong Ren

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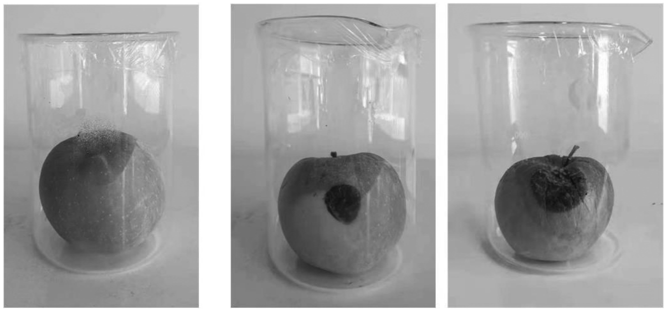
1. Introduction

Apple aroma is quite complicated, which comprises methanol, ethanol, acetaldehyde and a variety of volatile acids and esters. In the meantime, the aromatic compounds of apples are highly associated with the storage time and degree of deterioration [1]. During storage, apple fruits gradually lose the water and nutrient contents, and eventually become limp and crimping during late storage period, reducing the values as a commodity [2], [3]. Traditional apple detection methods, such as gas chromatographic method, gas chromatography–mass spectrometry method and liquid chromatography–mass spectrometry method, are expensive and time demanding, and may damage the integrity of specimens. Therefore, it is of great value to develop a device that can rapidly and non-destructively detect deteriorated refrigerated-stored apples to identify deterioration in early stages and reduce losses.

At present, the rapid development of testing technology is the non-destructive testing technology, mainly including spectral analysis technique [4], [5], machine vision technique and electronic nose technique. As an emerging rapid non-destructive detection and analysis technique on the basis of bionic olfactory principles [6], the electronic nose technique is a bionic system that comprises specific gas sensor arrays and a pattern recognition system. Presently, there has been a large body of research on fruit detection on the basis of electronic nose technology. Researchers usually study fruit hardness, sugar degree, decay, freshness and infectious diseases of bananas [7], oranges [8], cherries [9] and apples [10]–[13] by the electronic nose. The studies prove that the applicability of electronic nose to apple quality detection, while most scholars have focused the studies on apples artificially [14] infected with pathogenic bacteria without giving much attention to naturally deteriorated apples in cold storage. Electronic noses can be classified into two types: the first type is universal electronic noses, which are mainly costly PEN3 [15], [16]; the second type is customized, special-purpose electronic nose. A survey can easily reveal that these electronic noses mainly



(a) (b)

Figure 1. Apples after 120 days: (a) non-deteriorated apple and (b) deteriorated apple.

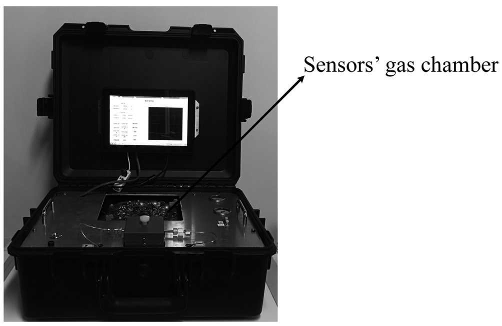


Figure 2. The electronic nose system.

comprise metal oxide sensor arrays [17]. In this paper, the sensor array comprises eight electrochemical sensors. Compared with metal oxide sensors, the "one-on-one" odour recognition feature of electrochemical sensors improves the precision and accuracy in specimens' odour detection.

In this paper, a proprietarily developed electronic nose system was utilized to detect whether apples have deteriorated in a temperature–humidity chamber to simulate the cold storage environment. Naturally deteriorated apples under a cold storage environment were used as research objects, sensor array comprises photoionization detector (PID) and electrochemical sensors, abnormal specimens were judged by two identical sensors, and data were analysed and processed by smoothing, PCA, kernel principal component analysis (KPCA), backpropagation (BP) and support vector machine (SVM) algorithms.

2. Materials and Methods

2.1 Experimental Materials

The "Fuji" apples used in this research are from an orchard based in Changping District, Beijing. Apples with similar degrees of ripeness and shapes that were free of diseases and mechanical damages on surfaces were selected. To simulate a cold storage environment, they were placed in a temperature–humidity chamber (4°C) for 120 days. After 120 days, as shown in Fig. 1, each specimen was taken out and placed in a 1,000 ml beaker, which was then sealed with sealing film and placed in a room temperature (25°C) environment for 30 min.

2.2 Electronic Nose Detection System

Figure 2 shows the electronic nose detection system, and Fig. 3 is the diagram of the electronic nose detection system. The electronic nose system mainly comprises the control unit, sensors' gas chamber, and data collection and transmission unit. The system has three electron magnetic valves, which are the sampling valve, feed valve and vacuum valve. The sampling valve controls gas entry; the feed valve controls the gas and its flow entering the gas chamber; and the vacuum valve controls the vacuum pump. The sampling and vacuum valves work collaboratively to prevent external gases from permeating into the instrument. The FameView [V7.6.12.4] is developed to collect output and save data generated by the electronic nose; data transmission and communication with the hardware component were supported by the MODBUS protocol to fulfil human–machine interaction. Data collected from the electronic nose were automatically saved by the system's software in the .csv format to facilitate offline analysis.

In this paper, two identical sensors (numbers 1 and 5) were selected as an indicator to discern whether a detected specimen is abnormal. If the numerical deviation of the two sensors is relatively large (higher than 1), the specimen would be deemed as abnormal and removed. Table 1 presents the sensor names and performance specifications. Table 2 presents specifications of common metal oxide sensors of PEN3 electronic nose. The electronic nose system designed in this paper comprises electrochemical

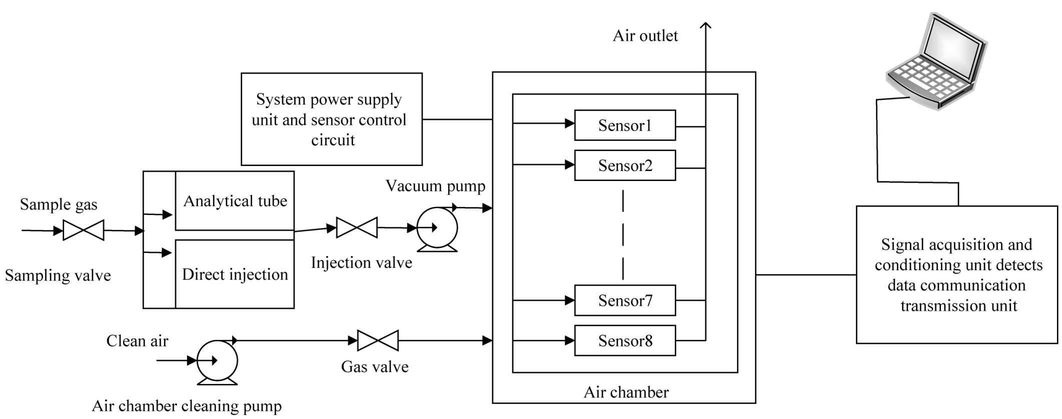


Figure 3. Schematic diagram of electronic nose detection system.

Table 1
Gas Sensors and Their Performance Specifications

Sensor	Sensor Model	Sensitive Gas	Detection Precision (ppm)
1	7NE/C_2H_4-20	C_2H_4	0.4
2	7NE/H_2S-50	H_2S	1
3	7NE/H_2S-1000	H_2S	20
4	7NE/C_2H_4-200	C_2H_4	4
5	7NE/C_2H_4-20	C_2H_4	0.4
6	7NE/ETO-20	C_2H_4O	0.4
7	7NE/PID-300	VOC	6
8	7NE/CH_2O-2000	CH_2O	40

and PID sensors. As can be known from Tables 1 and 2, compared with metal oxide sensors, electrochemical sensors have a higher precision for odour recognition.

2.3 Data Processing Method

2.3.1 Electronic Nose Data Acquisition

Prior to detection, the apples were taken out from the constant temperature–humidity chamber, then sealed and placed in room temperature for 30 min. To ensure there were no residual gases in sensors, the initial states were restored; the electronic nose was flushed with purified air for 20 min. After that, a sample needle was used to pierce through the sealing film to collect and save data of apple odours. The duration of the electronic nose's data collection was 7 min, the data sampling interval was 1 s and the gas flow rate was 150 ml/min.

2.3.2 Data Pre-processing

As gas sensors could be impacted by random noises caused by ambient temperature, humidity, vibration and flow disturbance, it is important to remove these signal noises. In this paper, three-point, five-point and seven-point linear smoothing algorithms were used for filtering and comparative analysis, as shown in Fig. 4. The results of the experiment indicate that the seven-point linear smoothing algorithm is better filtering result.

2.4 Evaluation Methods

Principal component analysis (PCA) is an effective method to extract eigenvalues in pattern recognition [18]. Drawing on the idea of dimensionality reduction, PCA uses a few variables to represent internal structures of multiple variables for them to retain as much information of the original variables as possible [19]. In this paper, the PCA

Table 2
Metal Oxide Sensors and Their Performance Specifications

Sensor Name	Sensitive Features	Representative Gas and Detection Precision ($\times 10^{-6}$ ml/m^3)
W1C	Sensitive to aromatic compounds	Methylbenzene, 10
W3C	Aromatic compounds, particularly sensitive to ammonia	Benzene, 10
W5C	Aromatic compounds like alkane and compounds with relatively small polarity	Propane, 1
W1S	Particularly sensitive to methane contained in specimens	Methane, 100
W2S	Particularly sensitive to ethanol contained in specimens	Carbon monoxide, 100
W3S	Sensitive to high concentration alkane, especially methane, in specimens	Methane, 100
W5S	Sensitive to nitrogen oxides, extremely sensitive to negative nitrogen oxides	Nitrogen dioxide, 1
W6S	Only detects hydrogen	Hydrogen, 100
W1W	Mainly sensitive to sulphides, also sensitive to organic sulphides	Hydrogen sulphide, 1
W2W	Mainly sensitive to aromatic compounds and organic sulphides	Hydrogen sulphide, 1

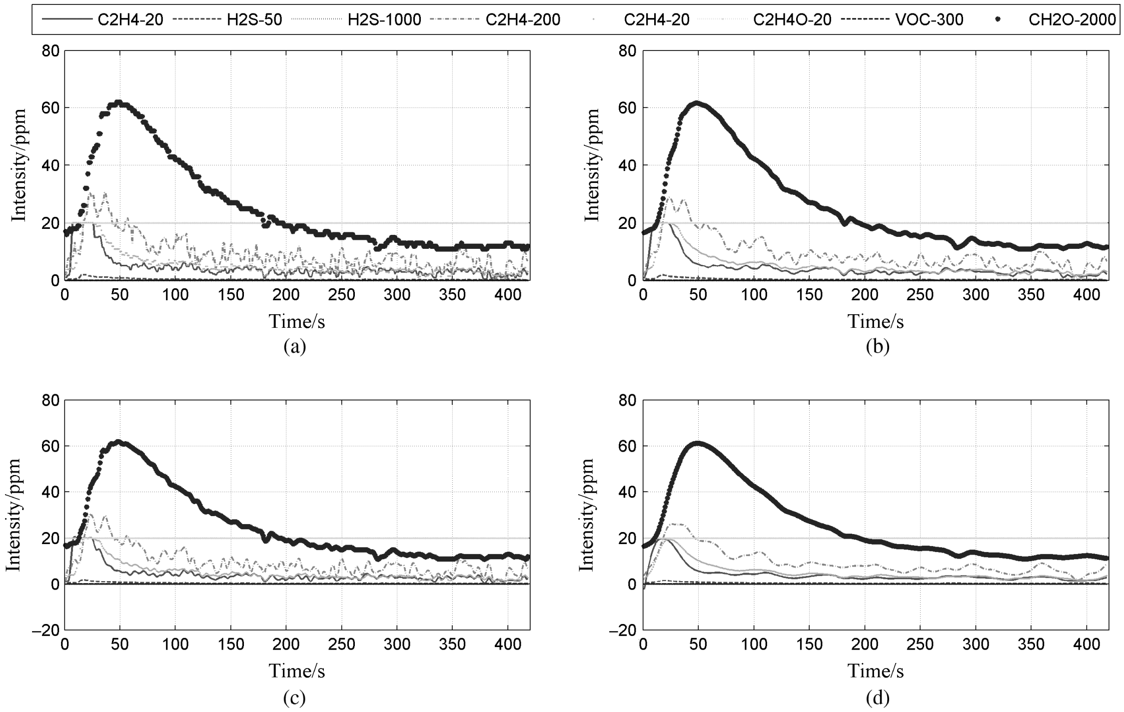


Figure 4. Original curve and filtered response curve: (a) sensor response curve; (b) three-point linear smoothing filter; (c) five-point linear smoothing filter; and (d) seven-point linear smoothing filter.

algorithm was developed on the basis of the eigenvalues-decomposition covariance matrix to rank the eigenvalues in a descending order.

KPCA is a non-linear extension of the PCA algorithm. The basic idea of KPCA is to map data from an original space to a high-dimensional space through incorporating a non-linear mapping function, and perform PCA on data in the high-dimensional space [20].

Backpropagation neural network (BPNN) constantly adjusts network parameters using gradient descent method by leveraging forward propagation of signals and backpropagation of errors, thereby getting the best approximation between the actual and expected network output values and achieving the purpose of training [21].

Machine learning algorithms are mainly divided into supervised learning, unsupervised learning and semi-supervised learning [22], [23]. SVM is a commonly used supervised machine learning algorithm, which is mainly used to solve classification and regression problems [24]. The non-linear relationships between data and eigenvalues of medium- or small-sized samples can be easily obtained using SVM. SVM is based on the principle of structural risk minimization, *i.e.*, to minimize both training and testing errors [25].

3. Results and Discussion

The experiment specimens are classified into two types (non-deteriorated apples and deteriorated apples). The total number of non-deteriorated and deteriorated apples from the same batch, with 20 for each type, was 40. Four abnormal specimens (deteriorated apples) were removed using the above-mentioned removal method.

3.1 Sensor Response and Feature Extraction

In combination with sensor response curve, an analysis of the collected data shows that sensor response tended to be stabilized after 200 s. Therefore, data generated between 200 and 350 s were selected as valid data in the analysis. To reduce the complexity of data to be analysed, the mean values of valid data collected from sensors in the stable states were used as eigenvalues to generate the graph of sensor responses.

Figure 5 shows the eigenvalues of eight sensors responding to the two groups of specimens. Among them, sensors numbering 1, 4 and 8 produced most pronounced responses, along with significant differences, to the two groups of specimens, indicating that these sensors were most sensitive to the odours of two groups of specimens and the volatile gases released by specimens were distinctly different.

PCA and KPCA analyses were conducted on electronic nose data generated by non-deteriorated apples and deteriorated apples, as shown in Fig. 6 (the asterisks represented deteriorated apples and the circles represented non-deteriorated apples). Figure 6(a) and (b) shows the results of PCA and KPCA analyses, respectively. In PCA analysis, the contribution rates of the first three principal components were 68.00%, 16.00% and 13.97%, with the total contribution rate achieving 97.97%; while the contribution rates of the first five principle components in the KPCA analysis were 43.54%, 16.59%, 12.35%, 7.78% and 6.19%, with the total contribution rate achieving 86.45%. As can be known from Fig. 6, satisfaction dimensionality reduction results were obtained for both PCA and KPCA, making them capable of differentiating deteriorated apples

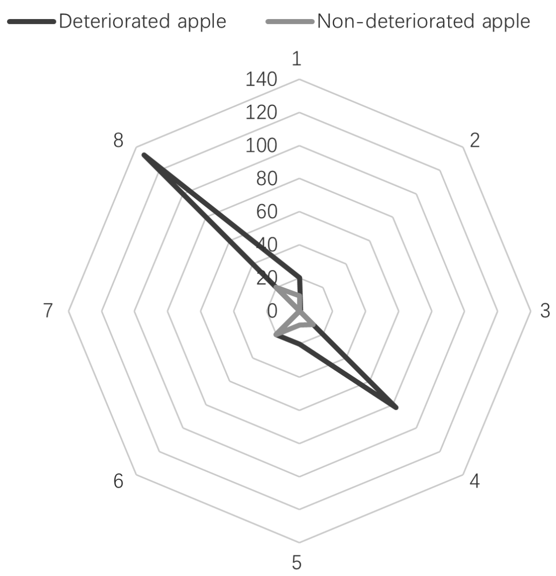


Figure 5. Sensor response radar graph.

from non-deteriorated ones. The same class was more clustered in the PCA. However, the total contribution rate of the first two principal components was lower in KPCA and dimensionality reduction result was better in PCA.

3.2 Identification Analysis

The leave-one-out cross-validation method was utilized to determine model predictions and estimates. Thirty-six specimens were divided into 36 subsets. Each specimen was used as a separate test set with the remaining 35 specimens as the training set; the procedure was repeated 36 times, deriving recognition accuracy rates of 36 models, the mean value of which was finally used as the recognition accuracy rate of the model.

Table 3
Recognition Results of Models

Recognition Method	TPR (%)	FPR (%)	F_1 score (%)	ACC (%)	Test Time of One Sample (s)
BP	93.75	5	93.75	94.44	0.019197
SVM	100	15	91.46	91.67	0.000256
PCA+SVM	100	5	96.97	97.22	0.00028
KPCA+SVM	100	15	91.43	91.67	0.001068

FPR, false positive rate.

When the model on the basis of BPNN was constructed, sensors' eigenvalues were used as network inputs to create a two-layer BP neural network where the hidden layer contains 25 nodes. The input data were pretreated with normalization, and then the newff function from the MATLAB toolbox was utilized to create the neural network. A Log-Sigmoid transfer function (logsig function) was used as the transfer function of the hidden layer; a Tan-Sigmoid function (tansig function) was used as the transfer function of the output layer; a momentum and self-adaptive lrBP gradient descent training function (traingdx) was employed to train the neural network; and a gradient descent momentum learning function (learngdm) was utilized as the learning function. The target error of network training was 0.02; the learning rate was 0.01; the additional momentum factor was 0.001; and the maximum training time was 1,000. For recognition results of the model, see Table 3.

Sensor eigenvalues, the first three principal component values of PCA after dimensionality reduction and the first five principal component values of KPCA after

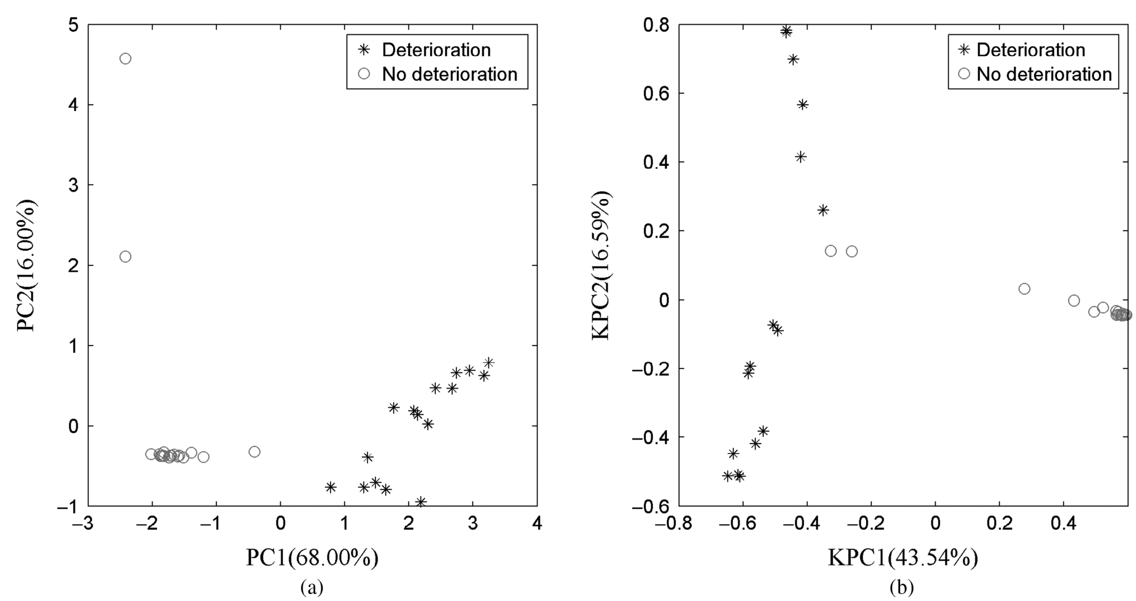


Figure 6. Feature extraction analysis: (a) PCA dimension reduction analysis and (b) KPCA dimension reduction analysis.

dimensionality reduction were used as input, and the types were used as the output to construct the model using MATLAB LibSVM toolbox. A radial basis function was used as the kernel function, where the penalty coefficient $C = 10$ and the regularization coefficient $\gamma = 0.1$.

To analyse the effectiveness of recognition algorithms of BP, SVM, PCA+SVM and KPCA+SVM towards non-deteriorated and deteriorated apple odours, Table 3 presents comparative results of these recognition methods.

The BP, SVM, PCA+SVM and KPCA+SVM models were utilized to discern and analyse deteriorated and non-deteriorated apples, yielding correct recognition rates of 94.44%, 91.67%, 97.22% and 91.67%, respectively. The true positive rate (TPR), F_1 score and accuracy (ACC) of PCA+SVM were higher than BP, SVM and KPCA+SVM, which were 100%, 96.97% and 97.22%. The test time of all algorithms for one sample (the mean test time of 36 samples) was less than 1 s, which meets the requirements of speed. The results show that the electronic nose has achieved fairly good recognition capability for deteriorated and non-deteriorated apples when working together with models constructed in this paper, and the recognition effect of PCA+SVM model is better than other models.

4. Conclusion

Reportedly, the majority of literature has focused on apples artificially infected with pathogenic bacteria. We select apples naturally deteriorated in the cold storage environment as research objects, which are more consistent with reality compared with earlier studies. In this paper, a proprietarily developed bionic olfaction-based portable electronic nose system was used to detect the occurrence of deterioration in apples. A smoothing pre-processing was first performed on the collected sample data, then the separability of non-deteriorated and deteriorated apples was discussed by drawing on PCA and KPCA algorithms, and finally the models on the basis of BP and SVM were constructed to analyse and discuss the feasibility to use electronic nose to rapidly determine non-deteriorated and deteriorated apples. The experiment results show that models on the basis of BP, SVM, PCA+SVM and KPCA+SVM reached a correct recognition rate up to 90% for deteriorated and non-deteriorated apples, but methods combining of PCA and SVM two algorithms produce better recognition result. This study only focuses on whether apples are inferior, but the degree of deterioration has not been studied.

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Biographies

Hui Tian received her master's degree in Beijing Information Science and Technology University. She is in the college of Automation at Beijing Information Science and Technology University. Her research interests include rapid detection of food and food safety.

Wenshen Jia received his Ph.D. degree in Jilin University. He is an associate research fellow in Department of Beijing Research Center for Agricultural Standards and Testing, Beijing, China. His research interests include modeling and instrument development.

Jie Ma received her Ph.D. degree in Harbin Engineering University. She is a Professor in Beijing Information Science and Technology University, Beijing, China. Her research interests include modelling and simulation.

Jihua Wang received his Ph.D. degree in China Agricultural University. He is a research fellow in Department of Beijing Research Center for Agricultural Standards and Testing, Beijing, China. His research interests include rapid detection of food and food safety.

Jianxiong Hao received his Ph.D. degree in China Agricultural University. He is a Professor in Hebei University of Science and Technology, Shijiazhuang, China. His research interests include rapid detection of food and food safety.