Available online www.jocpr.com

Journal of Chemical and Pharmaceutical Research, 2013, 5(9):575-583

Research Article

ISSN : 0975-7384
CODEN(USA) : JCPRC5

A comparison study of three nonlinear multivariate data analysis methods in smartongue: Kernel PCA, LLE and Sammon Mapping

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ABSTRACT

Smartongue is a voltammetric electronic tongue, based on a non-specific sensors array and one special voltammetry, so called multi-frequency large amplitude pulse voltammetry (MLAPV), which made its responding signals have much overlapping information. Three non-linear multivariate data analysis methods, Kernel principal component analysis (Kernel PCA), Locally linear Embedding (LLE) and Sammon mapping, were used to dig the information from the collecting data of Smartongue. One linear data analysis method, normal principal component analysis (PCA) and the discrimination index (DI value) were applied as the reference method and as the quantitative indicator to evaluate the discrimination ability. The results indicated that three non-linear data processing methods exhibited much more feasible and efficient than PCA in Smartongue. Sammon mapping is the most suitable non-linear method to process data in Smartongue. It was able to extract the useful information from the raw data and to classify three bitter solutions, six artificial green tea products and five milk powder solutions by means of the storage time well. Sammon mapping will be a very promising data processing technique for voltammetry electronic tongue.

Keywords: Smartongue; Kernel principal component analysis (Kernel PCA); Locally Linear Embedding (LLE); Sammon mapping

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INTRODUCTION

Electronic tongue [1] is a modern qualitative and quantitative analytical instrument with a cross-sensitive sensor array and appropriate pattern recognition for aqueous solution analysis. From the 1980s the first electronic tongue invented, many efforts have been invested to improve the feasibility and efficiency of electronic tongue. As a result, several types of electronic tongue were developed, for example, the potential electronic tongue by Toko research group [2] of Kyushu University in Japan and Legin research group [3] of Saint Petersburg University in Russia respectively, the voltammetry electronic tongue by Winquist research group [4] of Linköping University in Sweden and Martínez-Máñez research group [5] in Spain respectively, the electronic tongue based on impedance spectra by Mattoso research group [6] in Brazil and the electronic tongue based on surface acoustic wave (SAW) by Cole research group of Warwick University in England [7] and so on. Compared to other analytical methods, i.e. HPLC and GC, the electronic tongue has a special property: it collects the global information of analyzed objects by its sensors array and the acquired data has lots of overlapping information due to the non-specific sensors of the sensors array [8]. So this device, especially the voltammetric electronic tongue, requests a complex mathematics method of multivariate data analysis (MVDA) in order to extract the useful information from the data. Principal component analysis (PCA) [9] is the most popular technique for electronic tongue data processing. Other MVDA methods, i.e. PLS or SIMCA, are also used to process the data with good results [10]. However most of these techniques have the linear limitation and sometimes are almost infeasible to extract the non-linear information from the raw data generated by non-specific sensor array of electronic tongue in some cases. In contrast, non-linear multivariate data analysis method is more suitable for this task. Unfortunately, so far there are only a few available non-linear multivariate data analysis methods for data process in electronic tongue [11].

In this paper, we compared the feasibility and efficiency of three non-linear multivariate data analysis methods, Kernel PCA with three kernel functions [12], Local Linear Embedding (LLE) [13] and Sammon mapping [14], in dealing with the data measured by a voltammetric electronic tongue, Smartongue. This device developed by our lab bases on multi-frequency large amplitude pulse voltammetry (MLAPV), the potential applied and current responding technology which will produce lots of non-linear information of analyzed solutions. The results show that all three non-linear multivariate data analysis methods have better discrimination ability than PCA does. Furthermore Sammon mapping is the best among all three above mentioned methods.

EXPERIMENTAL SECTION

Samples Preparations and Measurements

Bitter solutions: Solutions of three bitter taste substances, caffeine (Sigma), acetaminophen (Sigma) and urea (Sigma), with analytical grade at 0.01 $mol \cdot L^{-1}$ were prepared respectively to evaluate the discrimination ability of Smartongue with three non-linear data process methods. Due to weak signal, the charging current and faradic current, generated by the multi-frequency large amplitude pulse voltammetry (MLAPV), bitter taste substance usually was not easily identified by Smartongue with normal PCA.

Green tea: One type of green tea, Longjing Tea, purchased from Lianhua Supermarket, Hangzhou, China, was used as the reference sample. In order to assess the ability of our device, the inferior green tea was produced by roasting the Longjing Tea in 120 °C for 5 min. Six green tea samples with the various mass ratios of Longjin Tea to inferior one were prepared to evaluate the discrimination ability of three non-linear multivariate data analysis methods (Table 1). In order to prepare tea solutions, 5 g of each sample had been brewed for 5 min in 250 mL deionized water (Milipore) at 85 °C with stirring. Then the tea-leave was taken away from solutions by the filter. All clear solutions were cooled down by ice-bath to 22 ◦C. 15 mL of the each solution was analyzed immediately after temperature got the setting point. Each sample equally divided into six pieces. Each piece was measured one time by Samrtongue.

The milk powder: One milk powder, Guangming milk powder, purchased from Lianhua Supermarket, Hangzhou, China, was used as the third sample for evaluation of the classification ability of three non-linear multivariate data analysis methods. In milk powder measurements, 25 g of Guangming milk powder was kept stirring in 250mL of deionized water (Millipore) at 40 °C until all powder was dissolved. Then the solution was stored in the refrigerator (Haier Co.) at 4 °C. Following the above mentioned method, five milk solutions were prepared on the same time of five successive days (Table 2). Before measurement, all solutions except the last one needed to be warmed up to 22 °C. Then 15 mL of the sample solution was analyzed immediately. Each sample equally divided into five pieces. Each piece was measured three times by Samrtongue.

Table 1. Six Green Tea Samples

Green tea	S1	S2	S3	S4	S5	S6
Total Weight (g)	5	5	5	5	5	5
Weight of inferior green tea (g)	0	1	2	3	4	5
Mass percentage of inferior green Tea (%)	0	20	40	60	80	100
Number of samples	6	6	6	6	6	6

Table 2. The Treatment of Milk Powder Solutions

Milk powder solutions	M5	M4	M3	M2	M1
Treatment Date	2011.04.01 8:00-11:00	2011.04.02 8:00-11:00	2011.04.03 8:00-11:00	2011.04.04 8:00-11:00	2011.04.05 8:00-11:00
Weight of Milk powder (g)	25	25	25	25	25
Volume of deionized water (mL)	250	250	250	250	250
Storage Time (Day)	4	3	2	1	0
Number of samples	5	5	5	5	5

Smartongue: Smartongue is a voltammetric electronic tongue developed by our lab (fig 1). As a standard three-electrode system, the device is consisted of six naked noble metal electrodes (diameter: 2 mm, Tianjing Aida Co., China), i.e. platinum, gold, palladium, tungsten, titanium, silver, as the working electrodes, an Ag/AgCl electrode (Tianjing Aida Co., China) as the reference electrode, and a platinum counter electrode (diameter: 2 mm, Tianjing Aida Co., China). Based on the signal excitation and responding method of multifrequency large amplitude pulse volatmmetry (MLAPV), this device gets a remarkable results to determine food properties, i.e. green tea, Chinese alcohol and modal medial [15] [16].

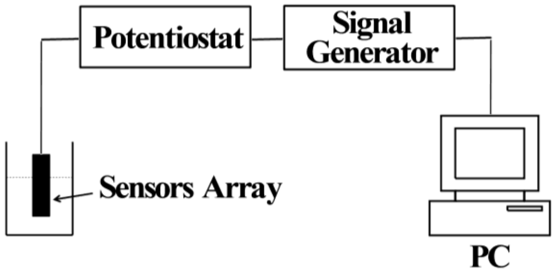

Fig. 1: Sketch map of the structure of Smartongue

Measurement principle: Multi-frequency large amplitude pulse voltammetry (MLAPV) with three frequencies, 1 Hz, 10 Hz and 100 Hz, was used by Smartongue (**Fig. 2**). The waveform of one frequency started at the pulse of the maximal voltage value at 1.0 V, and followed up pulses with the decreased amplitude rate at 0.2 V for each step till the minimal voltage value at −1.0 V. The interval between different successive frequency is 5 s with 0 V. According to the principle of MLAPV and the characteristics of non-specific sensor array of Smartongue, the data collected by Samrtongue contains abundant of overlapping and non-linear information [15] [16].

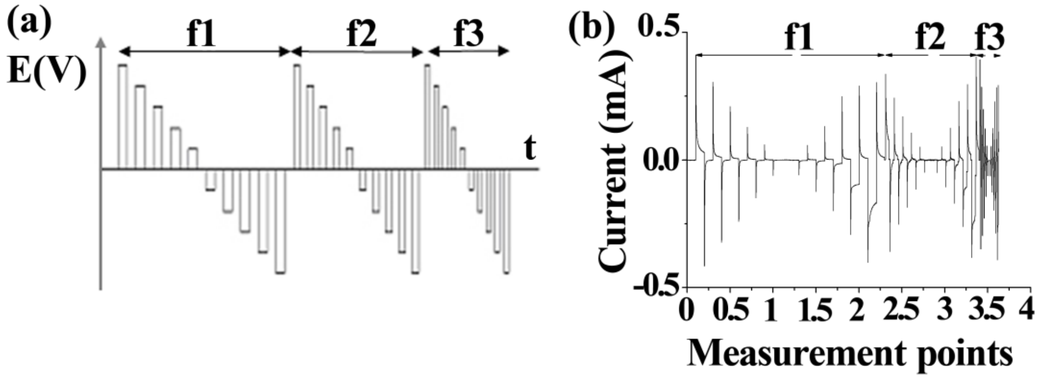

Fig. 2: Multifrequency large amplitude pulse voltammetry: (a) applied potential and (b) responding current of one working electrode

DATA PROCESSING

The data generated by six work electrodes in Smartongue was treated as input vectors. Three non-linear dimensionality reduction methods, Kernel PCA, LLE and Sammon mapping, were applied while one linear dimensionality reduction method, PCA, was used as a reference method. Discrimination Index (DI value) was used as a quantitative index for discrimination ability. All the methods were run by Matlab 2009b.

Principal Component Analysis (PCA): PCA (Principal Component Analysis) [9] is a traditional linear technique of dimensionality reduction. It transforms complex variables into some principal components by using dimensionality reduction technique, of which principal components are orthogonal and reflect main information of original variables. So far, it is the most commonly linear multivariate data analysis method for E-tongue and was used as a reference method in this paper.

Matrix X is assumed as the original data with n rows (samples) and p columns (eigenvalues). In principle, X is able to decomposed into two matrices

$$X = TL'$$

here matrix T is the score matrix with n rows and d columns (equal to the number of the principal components), which was used to exhibit the difference of the samples; matrix L is loading matrix with p rows and d columns. The diagonal elements of the matrix T'T are eigenvalues.

Kernel Principal Component Analysis (Kernel PCA): Kernel PCA is the reformulation of traditional linear PCA constructed by a kernel function [17]. The kernel function is a dot product of vectors of two samples in the feature space (a high dimensionality), and can be expressed by any function with a positive-semidefinite kernel K. It is not necessary to directly implement nonlinear mapping (called Φ) of the sample. Usually the kernel matrix K of the dataset X_i is defined by

$$k_{ij} = k(x_i, x_j)$$

where *k* is a kernel function. Now there are three common kernel functions, gauss kernel function, polynomial kernel function and sigmoid kernel function with the expressions as following:

Gauss - $k(x_i, x_j)=\exp\{-||x_i-x_j||^2/(2*\sigma)^2\}$

Poly - $k(x_i, x_j) = (x_i^T x_j + c)^d$

Sigmoid - $k(x_i, x_j) = \tanh(a x_i^T x_j + r)$

where σ, c and d, as well as r and a are constant parameters in each kernel function respectively. In the paper, all kernel functions are expressed by *K* without further differentiation in the Kernel PCA.

Local Linear Embedding (LLE): LLE used the local linear expression to mimic the nonlinear behavior. Thus the global structural information was provided by overlapping local neighborhoods without changing the whole geometric property during this process. The key point of LLE is to construct reconstruction weights. Generally, LLE algorithm is summarized by three steps:

(i) Construct a local neighborhood and select local neighbor points (k - nearest neighbor method) [18].
(ii) Calculate the reconstruction weights W_{ij} of samples to minimize the reconstruction error with one constraint

$$\min \varepsilon(W) = \sum_i || x_i - \sum_j W_{ij} x_{ij} ||^2$$

$$\sum_{j=1}^{k} w_{ij} = 1$$

where x_{ij} ($j=1,2,\ldots,k$) is *k* neighbor points of x_i, w_{ij} is the weight between x_i and x_{ij}.
(iii) Find Y_i that was embedded in low-dimensionality space of x_i by remaining w_{ij}. In order to fix Y and avoid collapsing to the origin of coordinates in low dimensionality, there are two constraints of Y: $\sum_{i=1}^{N} y_i = 0$, $\frac{1}{N}\sum_{i=1}^{N} y_i y_j^T = I$, where *I* is a N-dimensional identity matrix. Furthermore, Y should satisfy the following requirtments

$$\min \Phi(Y) = \sum_{i=1}^{N} || Y I_i - Y W_i ||^2 = \sum_{i=1}^{N} || Y(I_i - W_i) ||^2 = \min trYMY^T \quad YY^T = I$$

where $M = (I-W)^T (I-W)$. Using the Lagrange operator method, the result is $MY^T=\lambda Y^T$, the j^{th} component embedded by the i^{th} sample is $\sqrt{\lambda_i} v_{ji}$, where v_{ji} is the i^{th} component of v_j.

Sammon mapping: Sammon mapping is a projection method for geometric image dimensionality reduction. This method is a nonlinear transform to display the structural information of the original data in low dimensionality directly and visually via an approximate image of interrelation between high-dimensional points [14].

Assuming there are n points dictated by x_i (i=1,2,…,n) in the D-dimensional space and n responses indicated by y_i (i=1,2,…,n) to x_i in the d-dimensional space(d < D, usually d=2 or d=3). Besides, d_{ij}^* is denoted the distance between x_i and x_j, while d_{ij} the distance between y_i and y_j. These distances are the Euclidean Distance. With the initial value randomly selecting for y_i, the distance d_{ij} is determined by the following formula

$$d_{ij} = \sqrt{\sum_{k=1}^{d} (y_{ik} - y_{jk})^2}$$

The intrinsic algorithm of Sammon mapping is to find a mapping:

f:$X \in R^D \to Y \in R^d \ (d < D)$

The mapping is calculated by applying the iterative gradient method to minimize the objective function, and the low-dimensional expression is obtained after mapping. One vital characteristic of Sammon mapping is the conservation of the distance between the data during the transformation between two spaces with the formula read as

$$E = \frac{1}{\sum_{i<j} d_{ij}^*} \sum_{i<j}^{n} \frac{(d_{ij}^* - d_{ij})^2}{d_{ij}^*}$$

Discrimination Index (DI value): DI value [19] is a number to evaluate the separation level for the above non-linear multivariate data analysis methods. Generally there are two kinds of conditions to determine DI value.

(i) Each area has been clearly separated,
DI= $(1- \sum S_i / S_{all}) \times 100\%$

where S_i is the area occupied by the one group of samples; S_{all} is the area occupied by the whole samples. Thus, the bigger DI value reflects the bigger distance between two groups of samples.

(ii) Overlap is exist,
DI= $-(\sum S_i / S_{all}) \times 100\%$

(iii) The area was calculated by the following steps
1> Connect two points in the plot by straight line.
2> Repeat the first step for all points.
3> Determinate the maximal area defined by the above mentioned lines. The area is S.
4> S_i is the area from the points made by the ith sample.
5> S_{all} is the area from the points made by the all samples.

The maximum DI value is 100%, indicating the best separation of the samples.

RESULTS AND DISCUSSION

Classification of bitter solutions: The results of three nonlinear methods for bitter solutions are shown in **Fig. 3**. Here, the two-dimensional data from the biggest contribution of the PCA are provided as the initial locative value in d-dimensional space and 500 is the number of iterative operation for Sammon mapping optimal parameters. For LLE, the value of 10 is the optimal neighbor number and 30 is the eigenvalue number. Three kernel functions, i.e. gauss kernel function, polynomial kernel function and sigmoid kernel function are applied by Kernel PCA. As we have mentioned that PCA is used as the reference method. Due to its weak capacity of non-linear information process, PCA in **Fig. 3(a)** can not differentiate three bitter solutions well with the DI value -47.2%. In fact the data collected by Smartongue with the voltammetry of MLAPV contain lots of non-linear information. In addition, the weak charge molecules of caffeine, acetaminophen and urea in solutions complicate the collected information. In contrast, all three non-linear methods present well separations (**Fig. 3(b-f)**). It hints that Smartongue with multi-frequency large amplitude pulse voltammetry (MLAPV) [15] [16] [20] is able to obtain the characteristic information of the bitter solutions via the responding current, which contains abundant useful non-linear information. The Sammon mapping shows the best classification property with the biggest DI value of 95.4%. The three kernel PCAs with the gauss kernel function, the polynomial kernel function and the sigmoid kernel function respectively exhibits the different separation abilities in bitter solutions. The Gauss kernel function is the best for the discrimination of three bitter solutions among all three kernel functions although all the three kernel functions can distinguish samples well. The DI value of the Gauss kernel function is 93.2% while the DI values of the Polynomial kernel function and of the sigmoid kernel function are 86.9% and 92.1% respectively (**Table 3**).

Table 3. The Results of Different Data Processing Methods for Bitter Solutions

Data processing methods	PCA	Kernel PCA			LLE	Sammon mapping
		Gauss	Poly	Sigmoid		
Classification	－	＋	＋	＋	＋	＋
DI value (%)	-47.2	93.2	86.9	92.1	90.8	95.4

('＋' means all samples were distinguished, '－' means some samples were overlapped)

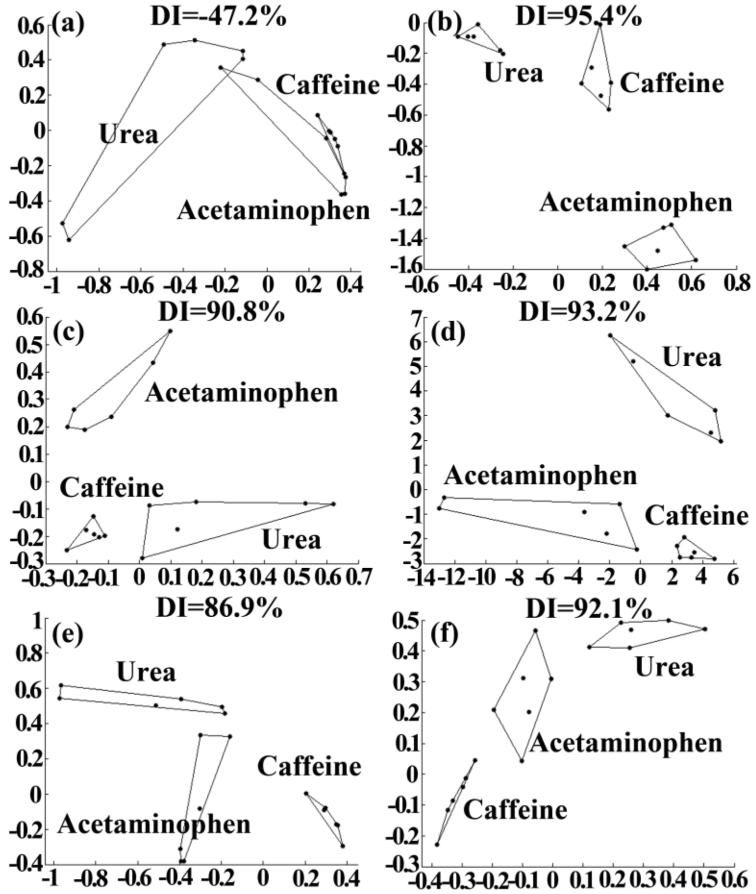

Fig. 3. The results of bitter solutions from the different data processing methods: (a) PCA; (b) Sammon mapping; (c) LLE; (d) Kernel PCA with gauss kernel function; (e) Kernel PCA with polynomial kernel function; (f) Kernel PCA with sigmoid kernel function

Separation of the artificial green tea samples: Fig 4 displays the results of three nonlinear dimensionality reduction methods in artificial green tea samples. The number of iterative operation of Sammon mapping is 500 and the initial locative value of green tea is provided by PCA. The optimal neighbor number is 10 and the optimal eigenvalue number is 30 for LLE. It can be seen that PCA has the worst separation ability among these project techniques due to its limited ability in non-linear data process. The six green tea samples cannot be discriminated by PCA and the DI value is -81.5% (**Fig. 4(a)**). In Kernel PCA treatments, only the Gauss kernel function can classify the quality of six green tea samples with the DI value 81.2%, while Kernel PCA with the polynomial kernel function and the sigmoid kernel function cannot differentiate six samples. The DI value is -34.6% for the polynomial kernel function and -78.4% for the sigmoid kernel function (**Fig. 4(d-f)**). Parallel Sammon mapping and LLE exhibits the strong separation ability, although the components of artificial green tea products were more complex than bitter solutions. Therefore they were more difficult for classification than the bitter solutions do. The DI value of Sammon mapping is 93.6% and the DI value of LLE is 89.7% (**see Fig. 4(b-c)**). All results for the DI value are summarized in **Table 4**. It is noticed that Sammon mapping shows the best discrimination ability for Smartongue with MLAPV.

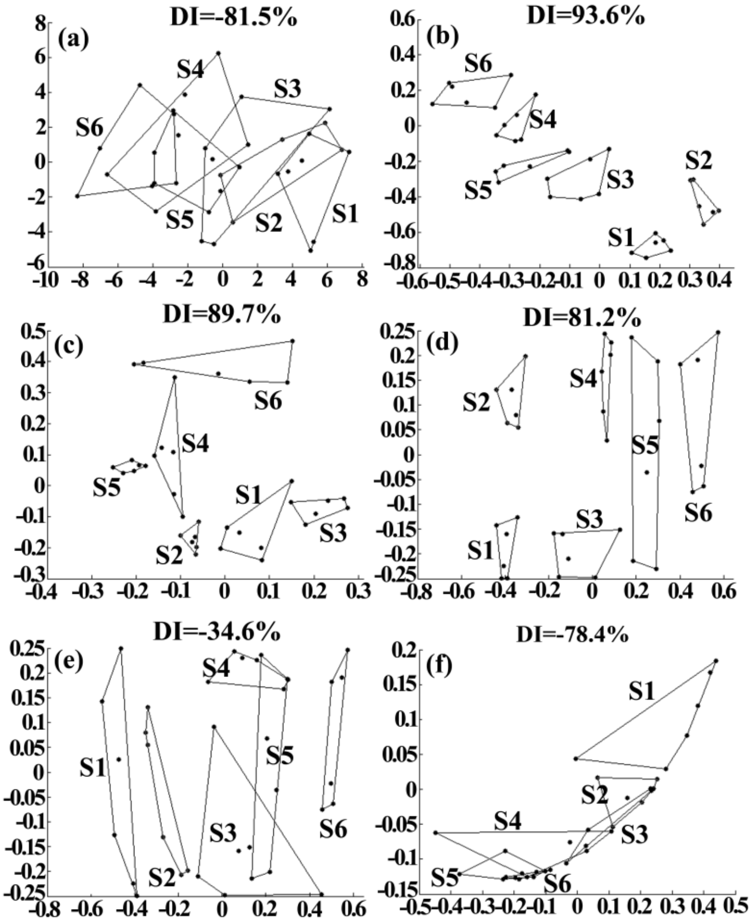

Fig. 4. The results of artificial green tea samples from the different data processing methods: (a) PCA; (b) Sammon mapping; (c) LLE; (d) Kernel PCA with gauss kernel function; (e) Kernel PCA with polynomial kernel function; (f) Kernel PCA with sigmoid kernel function

Table 4. The Results of Different Data Processing Methods for Artificial Green Tea Samples

Data processing methods	PCA	Kernel PCA			LLE	Sammon mapping
		Gauss	Poly	Sigmoid		
Classification	－	＋	－	－	＋	＋
DI value (%)	-81.5	81.2	-34.6	-78.4	89.7	93.6

('＋' means all samples were distinguished, '－' means some samples were overlapped)

Discrimination of the quality of milk powder with the different storage times: The quality changes of the milk powder solutions stored in 4 °C are assessed by Smartongue with four data processing methods (**Fig. 5**). The optimal parameters of Sammon mapping and LLE are same as the ones of the bitter solutions and artificial green tea samples. PCA still cannot differentiate the quality change of milk powder solutions in different storage times (**Fig. 5(a)**). The area of sample with 0 day storage time (Labeled M1) overlaps the one of sample with 1 day storage time (Labeled M2), although the area of other milk powder solutions with different storage time locates on different positions respectively. This is because the quality of milk powder downgrades with the storage time increasing due to its self enzymolysis and the microorganism from environment. But the change of quality in the first one or two days is slight with the evidence of the result of Kernel PCA with the polynomial kernel function (**Fig. 5(e)**). The signal of milk powder solution of 0 day storage time (Labeled M1) overlaps with the one of sample of 1 day storage time (Labeled M2), although the plot of Kernel PCA with the polynomial kernel function has a little better separation property than the plot of PCA does due to its non-linear data process ability. The DI value of Kernel PCA with the polynomial kernel function is -10.7% while

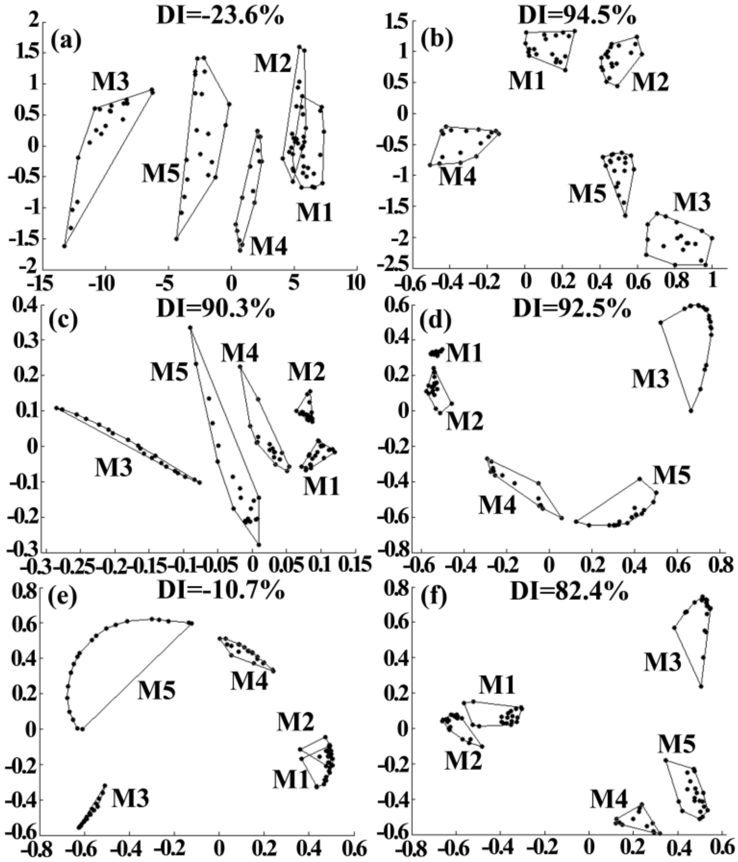

Fig. 5: The results of milk powder solutions from the different data processing methods: (a) PCA; (b) Sammon mapping; (c) LLE; (d) Kernel PCA with gauss kernel function; (e) Kernel PCA with polynomial kernel function; (f) Kernel PCA with sigmoid kernel function.

Table 5. The Results of Different Data Processing Methods for Milk Power

Data processing methods	PCA	Kernel PCA			LLE	Sammon mapping
		Gauss	Poly	Sigmoid		
Classification	－	＋	－	＋	＋	＋
DI value (%)	-23.6	92.9	-10.7	82.4	90.3	94.5

(‘＋’means all samples were distinguished, ‘－’means some samples were overlapped)

the DI value of PCA is -23.6%. The other non-linear techniques, i.e. Sammon mapping, LLE, Kernel PCA with the gauss kernel function, and Kernel PCA with the sigmoid kernel function, show an well separation ability for the quality change of milk powder solutions (**Fig. 5(b-d, f)**). The DI values of each non-linear method are listed in **Table 5**. The DI value of Sammon mapping is 94.5%, the highest value among all the non-linear data processing techniques.

CONCLUSION

In this paper, three kinds of non-linear multivariate data analysis methods, such as Kernel PCA with three kernel functions, Locally Linear Embedding (LLE) and Sammon mapping are applied to process the data from Smartongue with MLAPV, and PCA is used as the reference method. Three bitter solutions, six artificial green tea products and five milk powder solutions with different storage times, which are not easily separated by voltammetric electronic tongue with normal PCA, are our samples to evaluate the discrimination ability of the above mentioned methods. Three non-linear multivariate data analysis methods exhibit better separation ability than PCA, and Sammon mapping has the best discrimination ability for Smartongue. It indicates that the data from the electronic tongue contains plenty of useful non-linear information due to the non-specific sensors array and the waveform of MLAPV [8] [15]. The non-linear data process methods will have a great development in electronic tongue.

Acknowledgments
The project is supported by the High-Tech Research and Development (863) Program (2011AA1008047), the key projects of science & technology in Haikou (20110001) and the research foundation of education department of zhejiang province (Y201327111).

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