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Multifrequency large amplitude pulse voltammetry: A novel electrochemical method for electronic tongue

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Abstract

A novel electrochemical method, multifrequency large amplitude pulse voltammetry (MLAPV) for electronic tongue was introduced in this paper. It was constructed by several large amplitude pulse voltammetry (LAPV) with deferent step length. The applied waveform of MLAPV comprises three individual frequency segments, 1 Hz, 10 Hz and 100 Hz. The electronic tongue based on MLAPV was constructed by several metallic working electrodes, such as platinum, gold, titanium, nickel, palladium, a Ag/AgCl reference electrode and a pillar platinum electrode as counter electrode for standard three-electrode systems. Principal component analysis (PCA), a kind of multivariate data analysis (MVDA) was used for processing the data from the electronic tongue. In the present study, six Chinese distilled spirits and seven Longjing teas were analyzed by the electronic tongue based on MLAPV and were successfully discriminated by the working electrodes at different frequency segments. The results showed that frequency segments on the working electrode played a key role in discriminating various samples. MLAPV made the same metallic working electrodes have different separation ability at each segments. Better discrimination ability can be achieved by the combination of working electrodes with specific frequency segments. The present work showed that MLAPV is a useful method to reveal the specific frequency segment for each working electrode in various analysis systems.

Keywords: Multifrequency large amplitude pulse voltammetry; Electronic tongue; Chinese distilled spirits; Longjing tea

1. Introduction

The analysis devices, such as “electronic nose” [1,2] and “electronic tongue” [3–5], or “taste sensor” [6,7] have taken the growing interest in lots of researchers during the past two decades with the development of computer technology. These devices have a same structure of three principal components, sensors array, the equipment of emitting and receiving signals and pattern recognition. There are several specific characters: (i) sensors array instead of single specific sensors were used to obtain integral information of analyzed solutions, which concluded sample quality, state, etc.; (ii) the sensors in sensors array are non-specific sensors with lots of overlapping information, replacing the specific sensors of the traditional multisensor system for multicomponent analysis [8–10]; (iii) the devices need a complex mathematics method of MVDA for extracting the useful information from the data of the sensors array; (iv) the devices are supposed to mimic human sense, but not as same as human sense absolutely.

In recent years, three types of devices called electronic tongue, or taste sensor had been developed, which are based on potential, impedance spectroscopy or voltammetry. The taste sensor based on potential, was first presented by Toko and coworkers [6]. It was composed of several kinds of lipid/PVC membranes for transforming the taste quality information, such as sweetness, bitterness, sourness, saltiness, or umami into electric signal. Different types of foods such as soy sauce, beer, coffee and mineral water were investigated by the taste sensor, and not only the differences in foods were identified but also the taste interactions, such as suppression effect were detected [7,11–14]. Another type of potential electronic tongue was presented by Legin and coworkers [8], which was configured by several non-specific sensors based on chalcogenides glasses as transducers. It has been used for the discrimination of lots of foodstuffs and the analysis of some specific ions, or species in the solutions [15–18]. The second kind of electronic tongue

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based on impedance spectroscopy was first described by Riul and coworks, in which the sensors were constructed by supramolecular thin films of conducting polymers with a lipid-like material and were analyzed with impedance spectroscopy. This kind of electronic tongue could distinguish different brands of beverages and detect the low levels of taste, inorganic containment in water and suppression effect of taste sense [19–21]. The third kind of electronic tongue based on voltammetry, was first designed by Winquist et al. [3]. It comprised several metallic electrodes (platinum, gold, palladium, iridium, rhenium and rhodium) as working electrode, an Ag/AgCl reference electrode and a stainless steel electrode as counter electrode for standard three-electrode systems. It has been successfully used to analyze milk, tea, juice, drinking water and mold growth in liquid media [22–25].

Among various electronic tongues, the voltammetry electronic tongue developed by Linköping University displayed good performance due to its sensitivity, versatility, simplicity and robustness. Three types of waveforms, such as large amplitude pulse voltammetry (LAPV), small amplitude pulse voltammetry (SAPV) and staircase voltammetry, had been extensively applied for the voltammetry electronic tongue [23]. To the best of our knowledge, the waveform of LAPV with changes in its step lengths for the analysis of the samples by the voltammetry electronic tongue has not ever been described. In this paper, we reported a new waveform, i.e., multifrequency large amplitude pulse voltammetry (MLAPV), which was a combination of three waveforms of LAPV with different step lengths, 1 s, 0.1 s and 0.01 s, respectively. It was called “multifrequency” because the frequency of the pulse potential step changed in each segment due to its different step lengths. The electronic tongue with working electrodes based on the MLAPV displayed a well distinguish ability in six Chinese distilled spirits and seven Longjing teas. Moreover, the results showed that the separation ability of a specific working electrode in different frequency segments of MLAPV was not same. The frequency segments on the working electrode played a key role for the discrimination of the samples. MLAPV enabled the same metallic working electrodes to have different separation ability at each segment. Moreover, better discrimination ability can be achieved by the combination of working electrodes and specific frequency segments.

2. Theoretical description of MLAPV

MLAPV is a pulse voltammetry which is composed of a series of individual waveforms of LAPV with different step lengths. It is called MLAPV, because the frequency of the pulse step of each waveform of LAPV is different due to their different step lengths in each LAPV. It could be divided into several segments of frequency of waveform of LAPV.

In treatments, the principle of the each segment of MLAPV was the same as that of LAPV. When a potential was stepped to a final potential after waiting for a fix period on the base potential (0 V), a sharp transient current would flow to the working electrode, which was from the Helmholz double layer formed by charged species and electroactive compounds next to the electrode surface oxidized or reduced. The current would then decay as the double layer capacitance was charged and electroactive compounds were consumed, until only the limited diffusion faradic current remained. When the potential was stepped back to its base value (0 V), a similar but opposite reactions occur. Although the waveforms of each segment of MLAPV were similar as that of LAPV, some differences still existed due to the increase of the frequency of the potential steps. For example, the MLAPV had three segments of 1 Hz, 10 Hz and 100 Hz, which were equal to the corresponding segments of LAPV with the step length of 1 s, 0.1 s and 0.01 s. The segment of 1 Hz of MLAPV was the same as normal LAPV, since the charging current of the double layer capacitance had decayed to zero and the electroactive compounds next to the electrode surface was oxidized or reduced completely in the step length of 1 s. Each cycle of potential step started from the equilibrium of the reaction of the last cycle of potential step. But for the segment of 10 Hz and 100 Hz, it is different from normal LAPV, because the charging current of the double layer capacitance did not decay to zero and the electroactive compounds next to the electrode surface was not oxidized or reduced completely in the step lengths of 0.1 s and 0.01 s. Each cycle of the potential pulse step started from the transient state of reaction process of the last cycle step in the segment of 10 Hz and 100 Hz of MLAPV, rather than from the equilibrium of the reaction of the last cycle. So, compared with the segment of 1 Hz, extra information would be obtained from the segment of 10 Hz and 100 Hz of MLAPV.

3. Experimental

3.1. The voltammetry electronic tongue

As shown in Fig. 1, the electronic tongue consisted of several different metallic disc electrodes (platinum, gold, palladium, titanium, nickel, etc.) as working electrodes, an Ag/AgCl electrode reference electrode (saturated KCl, diameter 2 mm), and platinum counter electrode with a length of 5 mm and a diameter of 1 mm for standard three-electrode systems. The metal wire that served as working electrodes had a diameter of 2 mm and a purity of 99.9%. All the electrodes were made by Tianjing Aida Co. Ltd., China. A device called “Multifrequency large amplitude pulse scanner, MLAPS” (built at the lab of Sensory Science at Zhejiang Gongshang University) was a potentiostat with six

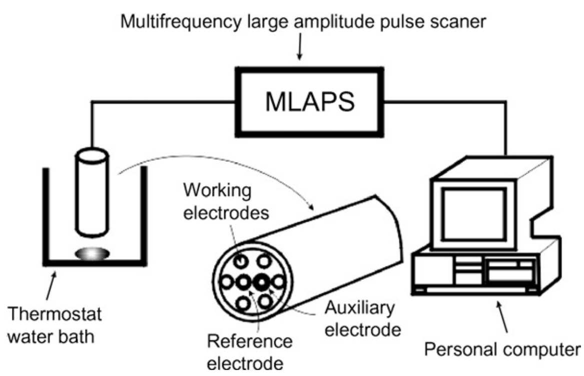

Fig. 1. The set-up of electronic tongue based on MLAPV.

Table 1
Chinese distilled spirits used in the measurements

Sample	Brand	Year	Flavor type	Ethanol degree (vol.%)	Amount (bottle)
A1	Niulangshan	2006	Fen-flavor	56	4
A2	Hongxin	2006	Fen-flavor	55	4
A3	Yili	2006	Luzhou-flavor	52	4
A4	Jianzhuang	2006	Luzhou-flavor	50	4
A5	Kongfujia	2006	Luzhou-flavor	44	4
A6	Shuanggou	2006	Luzhou-flavor	42	4

channels which was controlled by a personal computer (PC). It could make the potential pulses/steps on the working electrodes and enable the working electrodes to work consecutively one by one at three-electrode configurations which were controlled by a relay in MLAPS. The PC was used to set and control the potential pulses, measure and store current responses. A thermostat water bath was also applied to make the cell in a constant temperature.

3.2. Treatment of the samples

3.2.1. Chinese distilled spirits

Six Chinese distilled spirits (Table 1) bought from Carrefour, a French supermarket in China were selected as samples to evaluate the electronic tongue based on MLAPV. For each brand of Chinese distilled spirits, four bottles of distilled spirits picked from the shelves in random were used.

All the distilled spirits were produced by six different Chinese companies in 2006. The distilled spirits were classified as fen-flavor and Luzhou-flavor in flavor type, two in the type of fen-flavor and four in the type of Luzhou-flavor.

3.2.2. Longjing tea

Seven types of Longjing teas (a kind of famous Chinese green tea) were chosen as the second samples for the electronic tongue (shown in Table 2). As we all know, the quality of tea is linked to the climate of the planting geographical areas and the season of tea-leaf plucked. So, the Longjing teas' leaves were plucked from seven areas and two seasons which were summer (after 21 June) and spring (before 20 April) (see Table 2). Seven different areas were classified into three zones. Shuangfeng, Meijiawu and Longjing is the one of zones in Hangzhou, Zhejiang province, China, where the climate are most suitable for Longjing tea, and the tea produced there is called the Westlake Longjing tea. Wengzhou, Qiandaohu and Lishui belong to another zone out of Hangzhou, while they are also parts of Zhejiang province of China. They were the second suitable places for producing Longjing tea, and the teas from there are called Zhejiang Longjing tea. Fujian is another province in China, where is the area of the last zone out of Zhejiang province, China. The tea produced there is also called Longjing tea for commercial purpose.

All samples of Longjing tea were stored at −20 °C before test.

Table 2
Longjing teas used in the measurements

Sample	Period	The source of tea-leaf	Year
L1	Summer	Shuangfeng	2005
L2	Summer	Meijiawu	2005
L3	Spring	Longjing	2005
L4	Spring	Wengzhou	2005
L5	Spring	Qiandaohu	2005
L6	Spring	Lishui	2005
L7	Spring	Fujian	2005

3.3. Principle and procedure in measurement

3.3.1. Voltammetry method

The voltammetry electronic tongue with several metallic working electrodes was used in the measurements as described in Section 3.1. A current was measured between the working electrode and the counter electrode when a voltage was applied over the working electrode and the reference electrode. The applied potential waveform was MLAPV which was consisted of three segments of 1 Hz, 10 Hz and 100 Hz (the corresponding step length of each segments of LAPV is 1 s, 0.1 s and 0.01 s, respectively) (Fig. 2). The step length of 0 V was set 5 s between each segment to eliminate the interaction of the segment next to each other. The waveform of each segment of LAPV had the maximal value at 1.0 V and the minimal value at −1.0 V. The amplitude of each pulse was 0.2 V. A step of 0 V was inserted immediately before and after the pulse.

3.3.2. Chinese distilled spirits measurements

For distilled spirits measurements, the electronic tongue as described in Section 3.1 was constructed by two working electrodes, platinum and gold working electrode. The MLAPS enabled the working electrodes to work consecutively one by one at three-electrode configurations which were controlled by a relay in MLAPS. The applied potential was MLAPV as described in Section 3.3.1.

Eighty milliliters of each sample was used for analysis by the electronic tongue at the temperature (22 ± 1 °C) in random. Each solution was analyzed successively for three times, and then averaged the data to one point in score plot. So there would be 24 (4 × 6) points in the score plot.

3.3.3. Longjing tea measurements

In tea measurements, 4 g of each sample was stirred in 200 mL of deionized water at 85 °C and brewed for 5 min, and then the tea-leaves were filtered off. After the solution was cooled in an

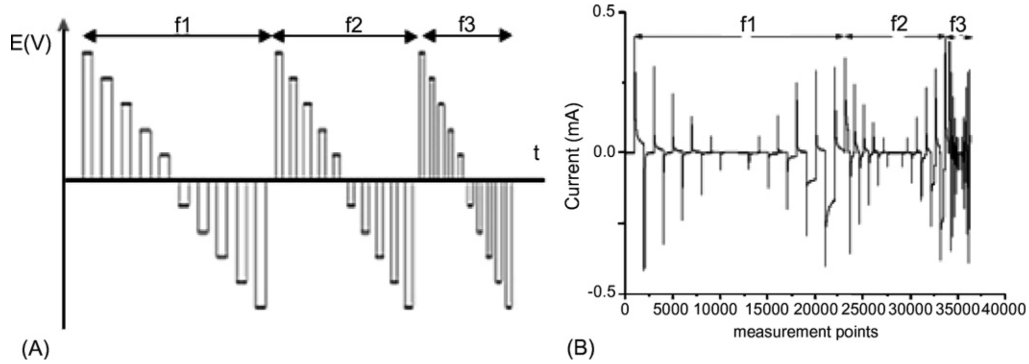

Fig. 2. Multifrequency large amplitude pulse voltammetry: (A) applied potential and (B) responding current of one working electrode.

ice-bath until the temperature dropped to 22 °C, 150 mL of the sample solution was analyzed immediately. Each of the seven tea samples was measured for four times in random, and each sample solution was analyzed three times successively. Then the data from three measurements were averaged to one point in score plot for the analyzed solution. So there would be 28 (4×7) points in the score plot.

The electronic tongue used in tea measurements was constructed by five metallic working electrodes, platinum, gold, palladium, titanium and nickel working electrodes. The MLAPS enabled the working electrodes to work consecutively one by one at three-electrode configurations which were controlled by a relay in MLAPS. Other components were the same as described in Section 3.1. The applied potential was MLAPV as described in Section 3.3.1.

3.3.4. Data processing

The multivariate data analysis (MVDA) has been extensively studied due to the rapid developments of the computer technology. Principal component analysis (PCA) is a popular and useful MVDA method for the analysis of the experimental data. PCA could decompose the experimental data matrix into latent variables and explain the variables in data by the loading plot from the loading vectors and the score plot from the score vectors. The loading plot could elucidate the relationships between the original variables and their influence on the system. The score plot displayed the relationships between the samples of the experiments, and could be used for grouping and classification the observations.

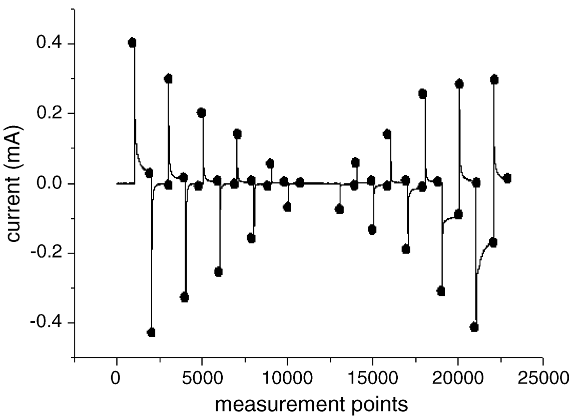

Fig. 3. The method of extracting data from the original data of one segment of the working electrode.

Since the experimental data were very large, a reasonable method of data processing was adopted that four points of each cycle which were related to the concentration and diffusion coefficients of the charged and electroactive compounds in the solution were collected (see Fig. 3). Then, there were 240 (Chinese distilled spirit) and 600 (Longjing tea) variables, respectively for PCA process according to the electronic tongue used in measurement described in Sections 3.3.2 and 3.3.3. As three different frequency segments, 1 Hz, 10 Hz and 100 Hz was processed by PCA, respectively, the data of one working electrode from the analyzed solutions would be divided into three independent data matrix by three frequency segments.

Principal component analysis (PCA) was performed with SPSS11.0 [26].

4. Results and discussion

4.1. Chinese distilled spirits treatments

4.1.1. The effect of each frequency segment on separation ability

The PCA score plots from the individual working electrodes of different frequency segments were shown in Fig. 4. It can be seen that each segment of frequency affected the separation ability obviously. Fig. 4(A–C) showed that the platinum electrode had different separation ability in three individual frequency segments for the discrimination of six Chinese distilled spirits. It differentiated all the six samples well in the frequency segment of 10 Hz (see Fig. 4(B)), while in 1 Hz, four distilled spirits, A1, A2, A3 and A4 were hardly separated (see Fig. 4(A)). Two samples, A1 and A2 were difficult to be separated in 100 Hz (see Fig. 4(C)). The result of the gold electrode was shown in Fig. 4(D–F). The only difference between the two working electrodes was that the gold electrode had the best separation ability in 100 Hz segment (see Fig. 4(F)), while for the platinum electrode, 10 Hz segment achieved the same goal. Two distilled spirits, A3 and A4, were not differentiated in Fig. 4(D), and four samples, A1, A2, A3 and A4, could not be distinguished in Fig. 4(E). The fact that MLAPV had different separation ability in its different frequency segments implied that the lengths

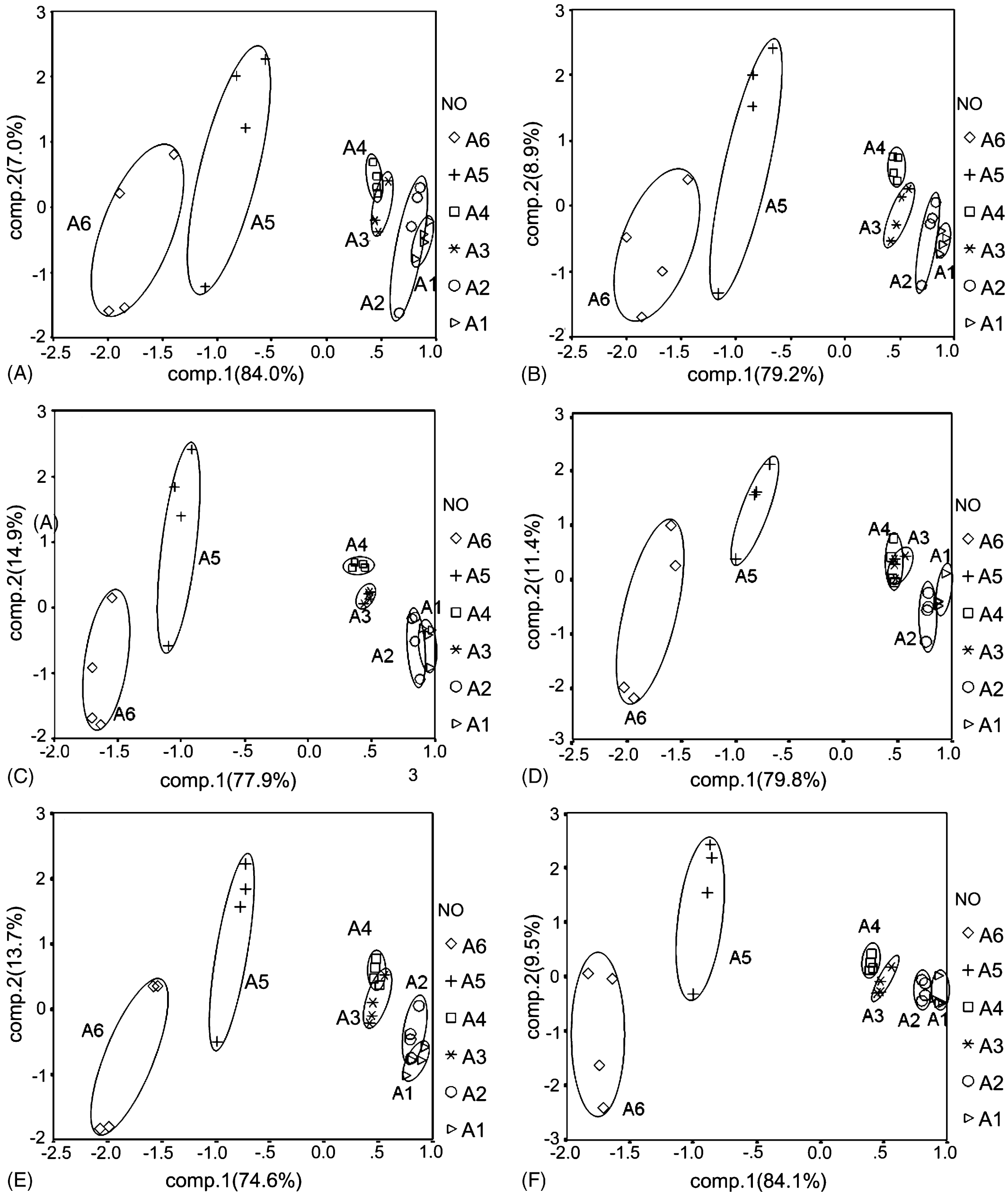

Fig. 4. The score plots of electronic tongue with platinum and gold electrode in different frequency: (A–C) platinum electrode was in 1 Hz, 10 Hz and 100 Hz and (D–F) gold electrode was in 1 Hz, 10 Hz and 100 Hz.

of the steps affected the discriminating ability of the working electrodes. The new information derived from the interaction between potential step cycle next to each other and obtained at the segment of 10 Hz and 100 Hz of MLAPV was useful in some cases, but the previous step cycle did not interact with the next step in the segment of 1 Hz of MLAPV due to its long step length of 1 s.

The MLAPV which was added a factor of frequency based on LAPV make it possible for the electronic tongue to discriminate samples with only one working electrode instead of sensors array. As was shown in Fig. 4(B) and (F), six different distilled spirits were well differentiated by the platinum and the gold electrode. The PC1 visualized the information of the ethanol concentration, and the concentration of the ethanol is gradually increased from the left to the right on the axis of comp.1.

4.1.2. The effect of MLAPV on separation ability

Fig. 4 showed the ability of the individual working electrode in three frequency segments of MLAPV for the discrimination of six distilled spirits, respectively. The well separation of the six distilled spirits can be seen from Fig. 4(B) and (F). To check the effect of MLAPV on the electronic tongue discriminating ability, the data of platinum electrode from the segment of 10 Hz and gold electrode in 100 Hz were merged, and a PCA was performed (see Fig. 5). Fig. 5 showed that the combination of two electrodes had better discrimination ability than by

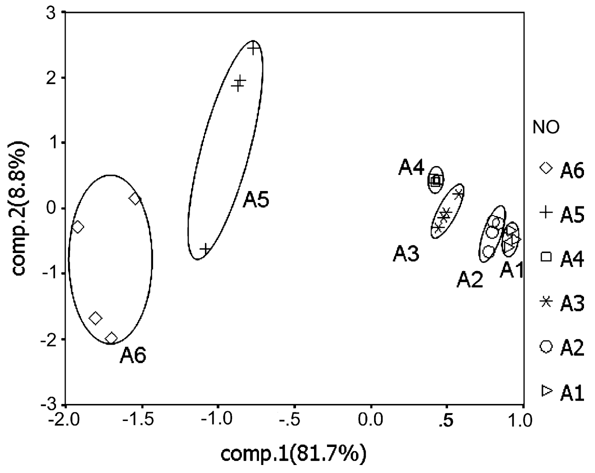

Fig. 5. The score plot from the merged data of the platinum electrode and gold electrode.

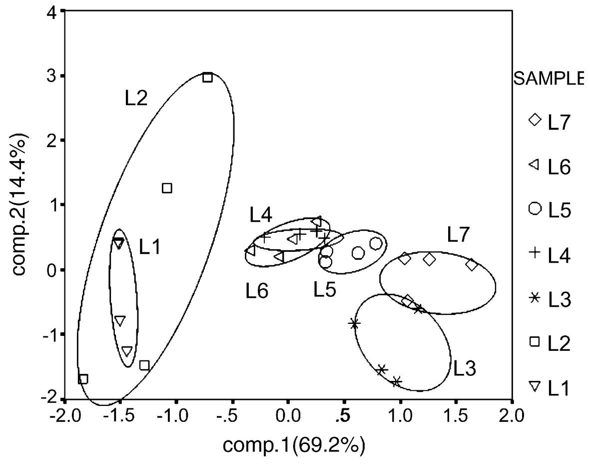

Fig. 6. The score plot from the 10 Hz segment of MLAPV.

each working electrode alone. The points of A4 had smaller dispersity and A2 and A1 were well separated. It could also be compared to other score plots of the merged data of platinum and gold electrode with different frequency segments (not shown), and the best discriminating ability was achieved in Fig. 5. Besides, the axis of comp.1 also reflects the content of ethanol of the samples and no more information could be derived from the score plots of PC1 versus PC2 and PC2 versus PC3 (not shown). The above result suggests that the data from the voltammetry electronic tongue based on MLAPV of each working electrode need not be collected all for PCA process. The data from each working electrode of their specific frequency segment of MLAPV should be merged as the valid data for PCA process.

4.2. Longjing tea treatments

4.2.1. Treatments as LAPV

To further prove the effectiveness of the MLAPV and to study whether the same working electrode need the same frequency segment of MLAPV for different sample measurements, Longjing teas whose tea-leaf were plucked from seven different places were analyzed by the electronic tongue with five metallic working electrodes such as platinum, gold, palladium, titanium and nickel. The MLAPV with three frequency segments, 1 Hz, 10 Hz and 100 Hz was also applied as described in Section 3.3.1. None of the PCA score plots could separate the seven Longjing teas as well as distilled spirits shown in Fig. 4(B and F) from one individual working electrode at each three frequency segment. It suggests that the seven Longjing teas are more difficult to be discriminated than six Chinese distilled spirits. Then the data as described in Section 3.3.4 from the 10 Hz segment of MLAPV of each working electrode were used for PCA just as the LAPV was applied with the step length of 0.1 s. The PCA score plot from the electronic tongue with five working electrodes was shown in Fig. 6. It visualized that the electronic tongue could not separate seven Longjing teas, with one L3 point close to the L7 cluster, L1 and L2, and L4 and L6 overlapped absolutely.

4.2.2. Treatments as MLAPV

The data from the 100 Hz segment of platinum electrode, 1 Hz segment of gold electrode, 1 Hz segment of palladium electrode, 100 Hz segment of titanium electrode and 100 Hz segment of nickel electrode were used for PCA score plots (see Fig. 7). Compared with Fig. 6, the MLAPV showed better discrimination ability than that of the LAPV. It can be seen from Fig. 7 that the MLAPV waveform can distinguish the summer tea from the spring tea by Line 1, with all the spring teas well separated. While for the two summer teas, L1 and L2, were hardly separated. The results implied that L1 and L2 had similar quality. The reason lies in that both Shuangfeng and Meijiawu where L1 and L2 were produced, respectively had the similar climate in Hangzhou, Zhejiang province. In addition, L1 and L2 were both produced in summer (see Table 2). It can also been seen that the spring teas, L3, L7 and three teas, L4, L5 and L6 had the difference quality apparently. The L3 cluster was far from the other spring tea cluster, and it could be separated easily with Line 2. In fact, the tea-leaf of L3 was from Longjing which is a famous interesting place of the West Lake for producing the best Longjing tea called West Lake Longjing tea. L7 was also far from the other clusters, while three clusters, L4, L5 and L6 were next to each other. It can be seen from Table 2 that the tea-leaf of

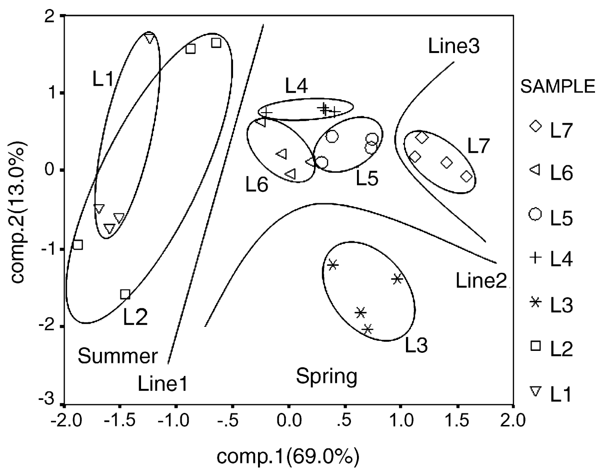

Fig. 7. The PCA score plot from the MLAPV.

L4, L5 and L6 were collected from the place out of Hangzhou, but in Zhejiang province. Three places, Wenzhou, Qiandaohu and Lishui had more similar climate and the tea-leaves from there were more suitable for Longjing tea than the tea-leaf of L7 from Fujian province, which is out of Zhejiang province. Three Longjing teas, L4, L5 and L6 were called Zhejiang Longjing tea, which was inferior to the West Lake Longjing tea but better than other Longjing tea. L7 had the worst quality due to its tea-leaf from the place out of Zhejiang province, where the climate was not suitable for Longjing tea.

The data from five working electrodes mentioned above with other frequency segments had also been collected for PCA process (not shown). Fig. 7 showed the best discrimination ability of electronic tongue. Also, the above results showed that different frequency segment is needed for one specific working electrode of MLAPV to discriminate different samples. The data from platinum working electrode and gold working electrode is more valid in the frequency segment of 100 Hz and 1 Hz of MLAPV for separating tea samples, respectively. While for Chinese distilled spirits, the data should be collected from the frequency segment of 10 Hz of platinum working electrode and the frequency of 100 Hz of gold working electrode.

5. Conclusions

In this paper, a novel voltammetry waveform, MLAPV which was used in electronic tongue has been described, and it has successfully classified six Chinese distilled spirits and seven Longjing teas well which were measured in random order. Also, the results showed that one working electrode had different separation ability in different frequency segments of MLAPV. Specific frequency segment on the working electrode plays a key role for the discrimination of various samples. To separate different types of samples, such as Chinese distilled spirits or Longjing tea by one specific working electrode, different frequency segments are needed. For example, in discriminating distilled spirits, 100 Hz segment of MLAPV and 10 Hz segment of MLAPV were chosen for the gold electrode and the platinum electrode, respectively, while 1 Hz segment of MLAPV was chosen for the gold electrode and 100 Hz segment of MLAPV was chosen for the platinum in separating Longjing teas.

In the present experiment, the segments of the MLAPV were set at 1 Hz, 10 Hz and 100 Hz. The frequency segments of MLAPV could be further developed, i.e., 1 kHz or higher, if the instrument is available. However, the increase of frequency of segment should be limited, since the applied potential would be linear weep when the frequency reached a specific value. The further work would be performed to find the highest frequency segment of MLAPV which is useful in voltammetry electronic tongue. The present study provides a useful method of MLAPV which reveal the specific frequency segment suitable for the working electrode for various analysis systems.

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