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Differentiating between ageing times of typical Chinese liquors by steady-state microelectrode voltammetry

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ABSTRACT

A typical Chinese liquor was characterised using a triple-electrode system with clean and simple design. In this approach, liquors were analysed with no other pre-treatment than mixing the liquor with a potassium chloride solution to ensure sufficient conductivity. Two broad reduction peaks at [−0.3 V, −0.5 V] and [−0.7 V, −0.8 V] were present in the cyclic voltammetry, demonstrating the feasibility of the gold electrode for liquor characterisation. Both the competing reactions in the liquor samples contributed to the reduction current. Thus, the steady-state microelectrode voltammetry affords an opportunity to estimate the total concentration of the redox-active species in the mixed compounds according to the relationship between the total concentration and the value of limiting diffusion current. Further, the discrimination of liquors at different ageing times was realised by the conjunction of the electrochemical results and principal component analysis.

1. Introduction

Wine is an alcoholic beverage consisting of hundreds of components that can influence the wine quality and flavour [1]. The understanding of wine chemistry centres on the sugar-containing plant tissue fermentation or biocatalysis, which involves redox reactions [2,3]. The phenolic compounds in wine phenolics are the initial substrates of oxidation, a fermentation process catalysed by metal ions, such as Fe^{3+} and Cu^{2+}, generating chemically active quinones and hydrogen peroxide [2]. These compounds promote the transformation of the sugar-containing plant tissue into an alcoholic beverage [4]. Usually, the identification and quantification of wine compounds are performed by means of analytical techniques, such as gas chromatography (GC) [5], mass spectrometry (MS) [6] and UV–visible spectrophotometry [7]. However, with wine chemistry challenges such as its complexity [8], considerable efforts may be expended before the various components are fully characterised [9]. In recent years, electrochemical techniques emerge as a promising strategy for the rapid, sensitive and economic analysis of foods and beverages when we consider the specific characteristics of conventional strategies, such long sample pre-treatment and analysis time, expensive equipment and the use of environmentally unfriendly reagents.

Considering that the key redox-active substances in wine can also be oxidised and reduced at the electrodes, electrochemical techniques have been applied to quantify these redox substances to explore the wine fermentation process [10,11]. Electrochemical methods, namely, linear sweep voltammetry (LSV), differential pulse voltammetry (DPV) and cyclic voltammetry (CV) using either carbon or metal electrodes, have been developed for the analysis of wine fermentation process [12–15]. A range of wine phenolics have been characterised by CV and used as basis for the effective analysis of polyphenols in wine [14]. Polyphenols are redox-active substances that are easily oxidised at carbon or metal electrodes. CV has been developed to discriminate red wines [16] and to quantify the total phenolic content in wine. Particularly, the CV curves are distinguished by resolving the whole peak into a set of peaks (peak-differentiating). As the peak-differentiating process is performed according to the electrochemical conversion, the final results of the peak analysis are represented as diagrams, called 'Redox Spectra of Wines' [17]. Electrochemical techniques have been regarded as promising methods for the identification and quantification of wine compounds.

The feasibility of electrochemical techniques has been proven, but they are still in the early stage of development [18,19]. The future direction of development aims to optimise the redox signal and improve

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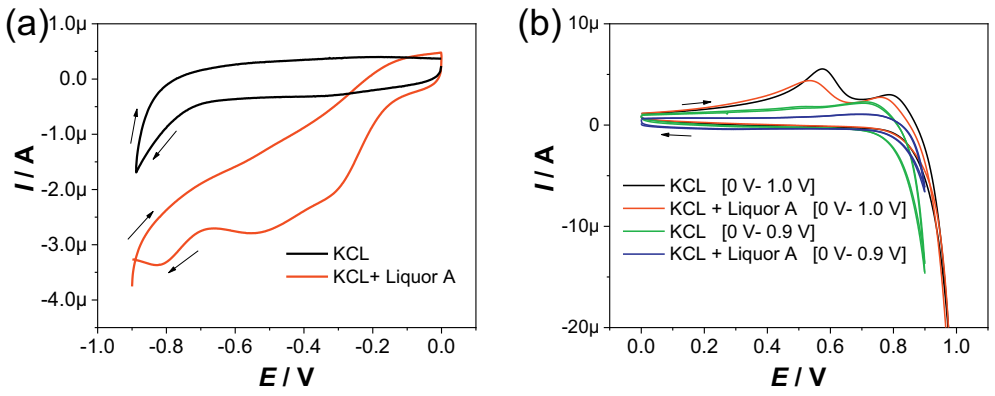

Fig. 1. (a) The cathode scanning of the cyclic voltammetry with a gold electrode in 0.1 M KCl (black line) and in 0.1 M KCl containing 5% liquor A (red line) respectively. Scan rate: 50 mV s^{-1}. (b) The anode scanning at [0 V, 1 V] in 0.1 M KCl (black line) and in 0.1 M KCl containing 5% liquor A (red line) respectively; the anode scanning at [0 V, 0.9 V] in 0.1 M KCl (green line) and in 0.1 M KCl containing 5% liquor A (blue line) respectively. Scan rate: 50 mV s^{-1}. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the precision of the relationship between the spectra and the components. In addition, current studies focus on low-alcohol grape wines, such as red and white wines [20,21]. In general, certain redox-active protective agents, such as ascorbic acid and SO_2, are added to prevent grape wine oxidation during storage in bottles, tanks or barrels; such redox-active agents may lead to interference signals in redox spectra recognition [3]. In this regard, electrochemical techniques may be more beneficial to the analysis of the Chinese liquor [22], which is produced without adding any redox-active agents. To the best of our knowledge, no research has used electrochemical techniques to predict the quality of Chinese liquor.

Different from low-alcohol grape wines, the Chinese liquor, which is produced without adding redox-active agents, generally contains a significantly higher alcohol content [23], and its components are fully exposed to atmospheric oxygen. Thus, the electrochemical redox signal obtained with a Chinese liquor sample may be more forthright and undisturbed compared with other alcoholic beverages. In this article, we present a proof-of-concept study combining steady-state voltammetry, UV–visible spectroscopy and principal component analysis (PCA) for the redox recognition of typical Chinese liquor with different ageing times. Our findings provide new insights into the chemistry of alcoholic beverages and help us understand how liquor ageing time affects liquor quality.

2. Experimental

2.1. Materials

Ethanol, ethyl acetate, ferrocenemethanol (abbreviated as FcMeOH) and KCl were purchased from Aladdin (Shanghai, China). All chemicals were used without further purification. All solutions were freshly prepared by ultrapure water, with a resistivity of 18.2 MΩ·cm^{-1}.

2.2. Liquor samples

Four Chinese rice liquor samples were collected from the manufacturer Xiangjiao Group Ltd., Shaoyang, China. The samples with different biological ageing times were named Liquor A (12 months), Liquor B (24 months), Liquor C (36 months) and Liquor D (48 months) respectively. All samples used in this study were directly collected from storage containers without any additive. The constituent analysis of liquor samples with different biological ageing times was primarily performed by gas chromatography (Agilent Technologies), as shown in Table S1 in Supporting Information. Model liquor is composed of the following main components: alcohol (64.0%, in volume ratio), ethyl acetate (276 mg/100 mL), ethyl butyrate (17 mg/100 mL), ethyl caproate (197 mg/100 mL), methyl alcohol (33 mg/100 mL), n-propanol (34 mg/100 mL), n-butyl alcohol (45 mg/100 mL), acetic acid (135 mg/100 mL), n-butyric acid (17 mg/100 mL), n-hexylic acid (58 mg/100 mL).

2.3. Methods

Electrochemistry. The polycrystalline gold electrode with the radius of 1 mm was polished with alumina slurries in different sizes (1, 0.3 and 0.05 mm) to achieve a mirror-like surface, followed by ultrasonic cleaning in ethanol and water. The microelectrode with the radius of 5 μm was polished with specialized polishing cloth, followed by ultrasonic cleaning in ethanol and water. Cyclic voltammetry was performed with a commercial electrochemical workstation (CHI660C) at room temperature near 298 K. For the three-electrode configuration system, a platinum wire was served as the counter electrode and a Ag/AgCl electrode served as the reference electrode. All the electrodes were purchased from CH Instruments, Shanghai, China. The oxygen was removed by passing of N_2 gas through the mixture before the recording of the voltammogram. The reported data for Cyclic voltammetry responses are the mean value of three parallel measurements. The error bars represent the standard deviation (SD) from the mean. UV–vis absorption spectra. The UV–vis spectra of liquor were determined by UV absorption spectrophotometry (SHIMADZU, UV-2450) at room temperature near 298 K. All the liquor samples are dissolved and diluted in 0.1 M KCl.

3. Results and discussion

Glassy carbon [16,24,25], platinum [26] and silver [27] electrodes had been tested for the voltammetric analysis of wines to improve their sensitivity or selectivity. In our study, the glassy carbon electrode was initially applied to monitor the electrochemical behaviour of liquor. We observed no evident oxidation peak using either LSV or CV (shown in Fig. S1 in Supporting Information). Afterwards, the polycrystalline gold disc electrode (1 mm in radius) was used to monitor the electrochemical behaviour of the liquor. To obtain the electrochemical information in the whole potential range [−1 V, 1 V] and avoid achieving an extremely large potential difference between the working and counter electrodes, cathode [0 V, −1 V] and anode [0 V, 1 V] scanning have been performed, respectively. Fig. 1a shows the cathode scanning of the CV. The CV scanned in 0.1 M KCl (as the control group) showed a non-peaked behaviour. This finding accords with the typical CV observations for polycrystalline gold disc electrode, indicating that the electrode surface is clean and stable [28]. The CV scanned in 0.1 M KCl containing 5% (volume ratio) liquor A presented two broad peaks: peak I at [−0.3 V, −0.5 V] and peak II at [−0.7 V, −0.8 V]. Such broad peaks hint that a wide range of substances in liquor A are redox-active and contribute to the total current. Fig. 1b illustrates the anode scanning [0 V, 1 V] of the CV. Unexpectedly, the CV scanned in 0.1 M KCl showed a peaked behaviour, which is similar with the CV scanned in 0.1 M KCl containing 5% (volume ratio) liquor A. In consideration of the oxidation process of gold, the peaks were caused by the adsorption of oxygen on the gold surface (equal to gold oxidation). The mechanism for gold oxidation has been demonstrated to proceed *via* a three-step

reaction sequence [29]. Although the oxygen in the electrolyte solution was cleaned by passing N_2 gas through the electrolyte solution before recording the voltammogram, traces of oxygen can be present in the electrolyte solution. To verify gold oxidation, anode scanning was performed in a lower potential [0 V, 0.9 V]. No peak was observed in the CV curves scanned in 0.1 M KCl or in 0.1 M KCl containing 5% liquor A. The oxidation peak disappeared in the condition of a lower potential, illustrating that the oxidation peaks observed in anode scanning [0 V, 1 V] were caused by oxygen adsorption. The oxidation peak observed in the CV of grape wine with a glassy carbon electrode was evident (with an oxidation peak close to 400 mV *vs.* Ag/AgCl) [12,16]; in this case, the CV peak for catechol or galloyl-containing phenolics oxidation in the samples can be 'covered' by the CV peak due to the strong adsorption of oxygen on the gold electrode. From the results of cathode and anode scanning, the gold electrode may be suitable for the electrochemical reduction (cathode scanning) rather than the oxidation (anode scanning) of mixed compounds in liquor.

The feasibility of gold electrode (1 mm in radius) has been proven. However, we observed that the correlation between the reduction current and liquor mass cannot be established due to the limited mass transport rate at the gold–electrolyte interface. As the mass transport rate to and from the electrodes increases along with the electrode size decreases [30], it is worth trying to utilize the gold microelectrode in the case of liquor compounds reduction. The gold microelectrode was characterised by steady-state voltammetry with the classical redox species ferrocene methanol (FcMeOH). Fig. 2a shows the steady-state voltammetry of 0.5 mM FcMeOH in 0.1 M KCl obtained with the microelectrode. The voltammetry of FcMeOH on the microelectrode formed an 'S' shaped curve. It started with a current close to zero and increased until it reached a steady-state, which resulted from the limited diffusion of FcMeOH to the microelectrode. A low value was observed with capacitative charging currents of the voltammetry. The results prove that the microelectrode was well prepared and satisfactory to subsequent experiments. Fig. 2b shows the CV of the microelectrode in 0.1 M KCl (black line, as control group) and in 0.1 M KCl containing 20% liquor A (red line). The CV curve of the control group (in 0.1 M KCl) indicates that almost no reduction current occurred in the considered potential range. On the contrary, the CV curve of the experimental group (in 0.1 M KCl containing 20% liquor A) showed an increased reduction current, which started at the potential of −0.38 V and reached a steady-state at the potential of −0.83 V.

Given the remarkable complexity of liquor composition, the reduction process is complicated by multiple competing reactions and is difficult to evaluate. However, both the competing reactions contribute to the reduction current. Thus, the microelectrode-based electrochemistry affords an opportunity to estimate the total concentration of the redox-active species in the mixed compounds. According to the formula of limiting diffusion current in a steady-state voltammetry [31,32]:

$$I_{plateau} = 4nFCDa \quad (1)$$

The total concentration of the redox-active species in liquor A was estimated to fall in the range between 1 mM to 8 mM, where n is the number of electrons involved in the electrochemical reaction. Here we assumed that the reduction of the mixed compounds proceeds *via* a single-electron transfer mechanism to estimate the total concentration of the redox-active species in the mixed compounds, with $n = 1$. The diffusion coefficient of redox species was estimated to fall in the range between $0.5 \times 10^{-6}\,cm^2\,s^{-1}$ and $3 \times 10^{-6}\,cm^2\,s^{-1}$, F is Faraday's constant, C is the bulk concentration of redox-active species and a is the electrode radius.

The steady-state potential (−0.83 V) is relatively negative and in such extreme conditions, the electrolytic system easily generates bubbles to interfere with the current of the working electrode. Therefore, we selected the half-wave potential −0.60 V to calibrate the concentration of liquor A. As shown in Fig. 3a, the reduction current at −0.60 V distinctly increased along with the increased concentration of liquor A, indicating the correlation between the reduction current and the liquor concentration. The dependence of the current response with the concentration was evident. Thus, the current is approximately the sum of different cathodic responses corresponding to non-interfering processes of the form $I = I_{plateau}$ x f(E), with $I_{plateau} = 4nFCDa$ and f(E) is a function of the applied potential. If the current measurements have been conducted at a constant potential, then f(E) is a constant. Therefore, the current at the half-wave potential (constant potential) can be reasonably used to calibrate the concentration of liquor A.

The reduction current at −0.18 V increased with the addition of liquor A. However, the increasing extent of the reduction current at −0.18 V was random and less than the reduction current at the half-wave potential of −0.60 V. The results indicate that the reduction of redox-active substances at a lower potential (less than −0.2 V) was interfered by the complicated liquor environment. Therefore, we concluded that the half-wave potential located in the middle of the reduction potential range [−0.38 V, −0.83 V] was suitable for calibrating the concentration of liquor A. Fig. 3b shows the CV of the microelectrode in 0.1 M KCl, 0.1 M KCl containing 20% C_2H_5OH, 0.1 M KCl containing 20% model liquor and 0.1 M KCl containing 20% liquor A in the control experiments. The artificial liquor model was mixed with various compounds according to the GC results of liquor A (shown in the Experimental section). The CVs recorded in 0.1 M KCl, 0.1 M KCl containing 20% C_2H_5OH, 0.1 M KCl containing 20% model liquor showed negligible reduction current, indicating that the microelectrode-based electrochemistry is effective for redox recognition of the liquor.

To verify the validity of the proposed method, the UV–visible spectroscopy of liquor A at different concentrations were measured. As shown in Fig. 3c, the absorbance at 205 and 277 nm significantly increased after adding liquor A. A UV absorbance at 205 nm usually indicates the availability of intermolecular or intramolecular conjugated charge transfer compounds, whereas that at 277 nm usually denotes the availability of benzene compounds and amino acids. Further, the UV–visible spectroscopy results were compared with the

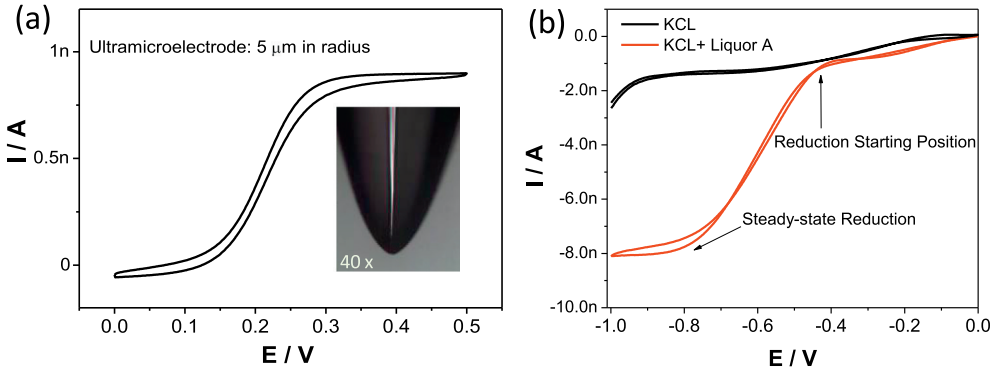

Fig. 2. (a) Steady-state voltammetry of 0.5 mM FcMeOH in 0.1 M KCl obtained at a microelectrode; radius, 5 μm; the insert is the optical photograph of the microelectrode. (b) The cyclic voltammetry of the microelectrode in 0.1 M KCl (black line) and in 0.1 M KCl containing 20% (volume ratio) liquor A (red line) respectively. Scan rate: 50 mV s^{-1}. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

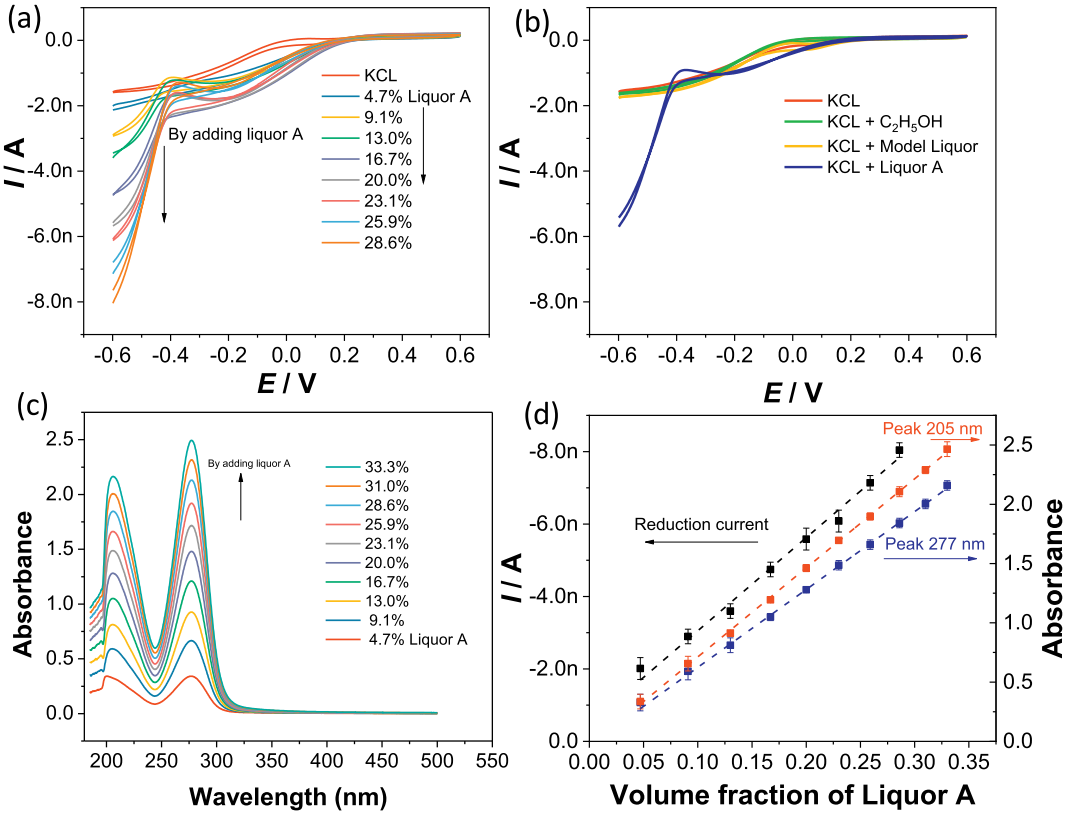

Fig. 3. (a) The cyclic voltammetry of the microelectrode in 0.1 M KCl and in 0.1 M KCl containing different concentrations of liquor A: volume ratio, 4.7%, 9.1%, 13.0%, 16.7%, 20.0%, 23.1%, 25.9%, 28.6%; scan rate: 50 mV s^{-1}. (b) The cyclic voltammetry of the microelectrode in 0.1 M KCl, 0.1 M KCl containing 20% C_2H_5OH, 0.1 M KCl containing 20% model liquor and 0.1 M KCl containing 20% liquor A respectively, scan rate: 50 mV s^{-1}. (c) The UV–vis spectra of liquor A at different concentrations: volume ratio 4.7%, 9.1%, 13.0%, 16.7%, 20.0%, 23.1%, 25.9%, 28.6%, 31.0%, 33.3%, all the samples are diluted in 0.1 M KCl. (d) Reduction current at half-wave potential −0.6 V (black spots) plotted *vs* volume of liquor A. Absorbance at 205 nm (red spots) and 277 nm (blue spots) plotted *vs* volume of liquor A. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

electrochemical results. Fig. 3d illustrates the plotted reduction current at −0.6 V plotted *vs* the volume of liquor A and the plot of peak absorbance *vs* the volume of liquor A. Both the reduction current and peak absorbance showed similar rising tendencies, suggesting the concentration-dependent behaviour of the reduction current. UV–visible spectroscopy is a standard method in monitoring spectral properties of organic compounds [33]. However, UV–visible absorption is inapplicable in characterising all liquor compounds. Thus, the application of UV–visible spectroscopy in liquor characterisation is limited. In this case, the conjunction of electrochemistry and other modern techniques gains importance in exploring the redox reactions occurring in liquor or wine fermentation process [34].

Regarding the chemistry of Chinese liquor, a close link exists between liquor ageing time and liquor quality [35]. Therefore, it is important to understand how liquor ageing time affects liquor chemistry. The constituent analysis of liquor samples with different biological ageing times was performed by GC. As shown in Fig. 4a, the main constituents for liquor samples with different biological ageing times are the same: short-chain fatty acids, saturated ethanols and esters. Studies have reported that polyphenols are redox-active substances that are easily oxidised at carbon or metal electrodes [14]. However, in the case of liquor, the cathodic current may result from the reduction of aldehydes [36], short-chain fatty acids (*e.g.* formic acid) [37] and alkyl esters [38]. For instance, the electrochemical reduction of acetaldehyde to ethanol on copper was demonstrated in a study combining online HPLC analysis and DFT calculations [39]. As depicted in Fig. 4b, in the process of acetaldehyde reduction, the protonation of the intermediate $CH_3CHO\cdot$ occurred at the potential of 0.45 V, which is close to the half-wave potential in the process of liquor reduction. In the liquor fermentation process, the short-chain compounds transform into long-chain molecules. The content of short-chain esters (*e.g.* ethyl acetate) in the 12-month samples (276 mg/100 mL) was significantly higher than that in the 48-month samples (176 mg/100 mL). On the contrary, the content of ethyl caproate in the 12-month samples (256.3 mg/100 mL) was significantly lower than that in the 48-month samples (437.7 mg/100 mL). The chromatography characterisation above-described enables us to confirm the main constituents in the liquor samples; however, it is not likely to provide further details in regard to the discrimination of liquors at different ageing times.

The CV of liquor samples with different biological ageing times was performed, and the reduction current and half-wave potential were plotted *vs* the samples, as shown in Fig. 4c. The reduction current at the half-wave potential slightly increased along with the prolonged ageing time, whereas the half-wave potential remained unchanged. In consideration of the standard deviation, the use of reduction current as the indicator of liquor ageing time was satisfactory. However, the reduction current resulted not from the action of a single factor (biological ageing time) but from the interaction and counteraction of the action system among the material, liquor manufacturing technology and liquor storage environment, *etc*. To address the challenge of liquor samples, PCA has been performed on the CV data. PCA is a scientific method of processing the correlation among different factors. As certain interaction and counteraction exist between indicators and targets, the complexity of analysing the correlation increases. PCA can reassemble the original indicators into newly comprehensive ones, which are uncorrelated and replace the original indicators.

In this case of CV data, the first principal component accounted for the vast majority of the total variance in the raw data. Thus, the

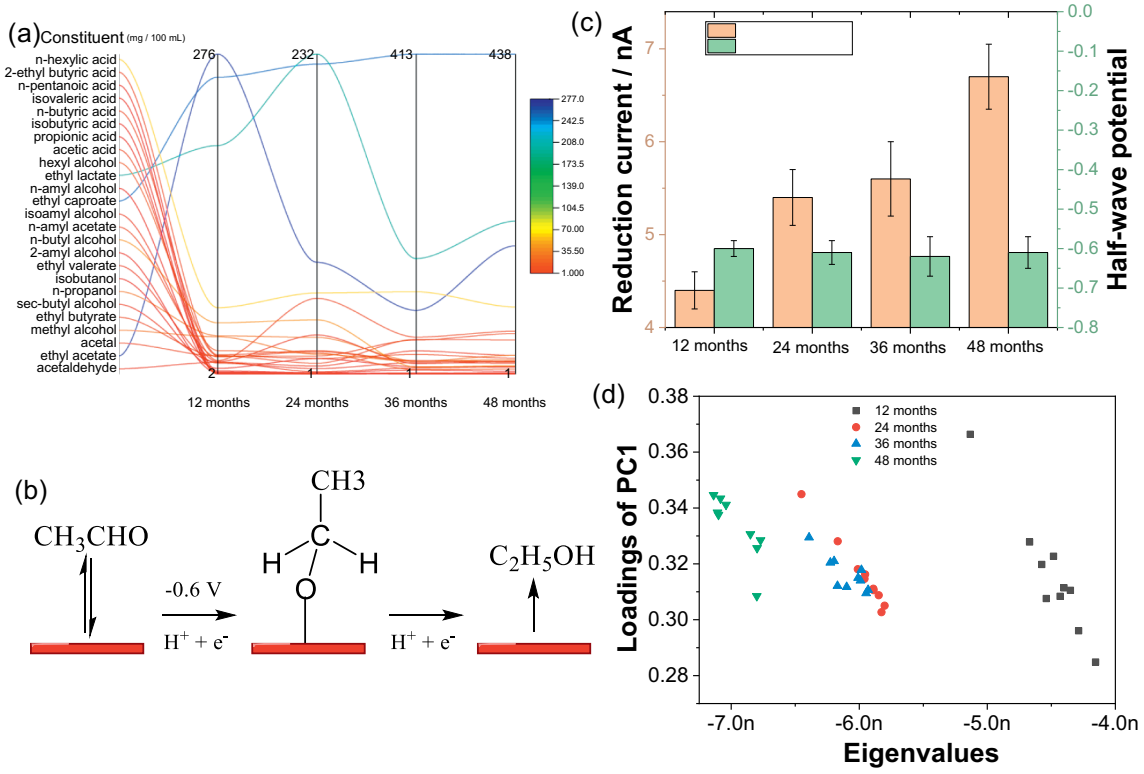

Fig. 4. (a) The constituent analysis of liquor samples with different biological ageing times performed by gas chromatography. (b) Mechanism of the electrochemical reduction of acetaldehyde on the gold electrode. (c) Reduction currents and half-wave potentials plotted *vs* liquor samples of different ageing time: 12 months, 24 months, 36 months and 48 months. The error bars represent the standard deviation (SD) from the mean. (d) PCA loading values of PC1 plotted *vs* eigenvalues.

combination of PC1 and eigenvalues was selected as the best model for the discrimination of liquor ageing time (Fig. 4d). Every dot in the PCA diagram refers to a CV curve obtained in our experiment. The dots in the PCA diagram showed varying populations because of the different component ratios for various samples. A general separation was observed between the liquor samples with different ageing times. The results indicate that discrimination among the ageing times of liquors is possible with the steady-state microelectrode voltammetry. Notably, the data regions for the samples with ageing time of 24 and 36 months are close and cannot be well distinguished. However, different reduction currents were observed for the two samples shown in Fig. 4c. The results indicate that the samples with ageing time of 24 and 36 months may be distinguished by building further classification models (to sharpen the separation between samples), and further research using partial least squares discriminant analysis is currently underway to shed more light on this topic. Similar works aimed at discrimination among wines using analytical techniques, such as electronic tongue, electronic nose, GC and MS, can be found in the literature [40–42]. For example, a technique combining the electronic tongue and PCA was employed to differentiate brandies; it allowed either the detection of the used alternative ageing practices or the assessment of a quality index of brandy samples [42]. These studies showed that the combined techniques offer a powerful potential in the classification of wines. These methods are often based on either modified electrodes or elaborate methodologies. In this study, on the one hand, the proposed method is based on the triple-electrode system with clean and simple design. On the other hand, from an analytical viewpoint, the main advantage of the proposed method is the possibility of direct analysis of the liquor sample without the need for pre-treatment or separation.

4. Conclusions

We have firstly proven the feasibility of using a gold electrode with 1 mm radius in monitoring the reduction reaction of typical Chinese liquors. However, we observed that the correlation between the reduction current and liquor mass cannot be established due to the limited mass transport rate at the gold–electrolyte interface. As the mass transport rate to and from the electrode increases along with the decrease in the electrode size, the microelectrode with a radius of 5 μm was used to afford a fast mass transport rate. Further, the correlation between the reduction current and liquor mass was established by the proposed microelectrode-based electrochemistry. Moreover, the discrimination of liquors at different ageing times was realised by the conjunction of steady-state microelectrode voltammetry and PCA. This study displayed that the steady-state microelectrode voltammetry as an alternative methodology is complementary to the conventional chromatographic and mass spectrographic characterisations and may be important for exploring the redox reactions occurring in the liquor fermentation process.

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Declaration of competing interest

There are no conflicts of interest to declare.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.microc.2019.104244.

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