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An electroanalytical method for brewing vinegar authentic identification

Un método electroanalítico para elaboración de identificación auténtica de vinagre

L. Fu[1,3*], H.Y. Zhang[1], Y.H. Zheng[2], H.W. Zhang[1], Q.H. Liu[3]

[1]*College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou, 310018, P.R. China.*
[2]*Institute of Botany, Jiangsu Province and Chinese Academy of Sciences (Nanjing Botanical Garden Mem. Sun Yat-Sen), Nanjing, 210014, PR China.*
[3]*Hongze Fishseeds Biotech, Inc. Hongze, Huaian, 223125, PR China.*

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Abstract

An electrochemical fingerprint-based methodology is described here for vinegar authentic identification. The commercial three-electrode system can be directly inserted into vinegar for voltammetric profile recording without the addition of electrolyte. The free amino acids and aroma compounds produced from the fermentation process can be oxidized during the differential pulse voltammetric scan. The fingerprint pattern of vinegar varies between the different brand due to the different raw ingredients and fermentation process. The DPV profiles can be converted into 2D scatter patterns for identifying twelve different vinegar in this work. In addition, clustering analysis confirmed the feasibility of the proposed method for vinegar authentic identification. We believe the proposed methodology can be further extended for other food quality screening application.

Keywords: electrochemistry; vinegar; glassy carbon electrode; cluster analysis; food safety.

Resumen

Aquí se describe una metodología electroquímica basada en huellas dactilares para la identificación auténtica de vinagre. El sistema comercial de tres electrodos se puede insertar directamente en vinagre para registrar el perfil voltamétrico sin la adición de electrolito. Los aminoácidos libres y los compuestos aromáticos producidos por el proceso de fermentación pueden oxidarse durante la exploración voltamétrica de pulso diferencial. El patrón de huella digital del vinagre varía entre las diferentes marcas debido a los diferentes ingredientes crudos y al proceso de fermentación. Los perfiles DPV se pueden convertir en patrones de dispersión 2D para identificar doce vinagres diferentes en este trabajo. Además, el análisis de agrupamiento confirmó la viabilidad del método propuesto para la identificación auténtica de vinagre. Creemos que la metodología propuesta puede extenderse aún más para otras aplicaciones de detección de calidad de alimentos.

Palabras clave: electroquímica; vinagre; electrodo de carbono vidrioso; análisis de conglomerados; seguridad alimenticia.

1 Introduction

Vinegar is a traditional fermented condiment widely used in cooking. It is a sour liquid condiment, mainly made by fermentation of sorghum, rice, corn, wheat and bran. According to the production process, vinegar can be divided into brewing vinegar and synthetic vinegar. Brewing vinegar is made from food, sugar and ethanol using microbial fermentation, while the synthetic vinegar is made by mixing of water, acid agent, seasoning, spice and food coloring with edible acetic acid. White vinegar, rice vinegar and mature vinegar are three main types of brewing vinegar. The main raw material of white vinegar is glutinous rice. The starch in glutinous rice is converted into sugar. Then yeast is used to ferment it into alcohol. Due to its light color, white vinegar is mainly used for mixing cold dishes and western food (Junqueira *et al*., 2019; Núñez-Gastélum *et al*., 2019). Rice vinegar is mainly made by fermenting rice and sorghum, which has a light taste. In contrast, the mature vinegar needs a long time in the brewing process, in which a small amount of alcohol reacts with organic acids to form aromatic substances, which has a stronger taste.

So far, a lot of work has focused on the

* *Corresponding author. E-mail*: fuli@hdu.edu.cn
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composition analysis of vinegar. For example, Mejías *et al*. (Castro Mejías *et al*., 2002) studied the aromatic compounds in vinegar using a headspace solid-phase microextraction process (Paéz-García and Valdés-Parada, 2019; Torres-Segundo *et al*., 2019). Gálvez *et al*. (Gálvez *et al*., 1994) used ethyl acetate for the extraction of the polyphenolic compounds. The compounds were subsequently separated and determined using HPLC. Thomas and co-workers demonstrated the analyses of acetic acid from vinegar using ^{2}H NMR and ^{13}C-IRMS (Thomas and Jamin, 2009). Although these methods could be used for the determination of quality and authenticity of vinegar, the complicated sample preparation process and expensive instruments restricted their field applications. Except for the experimental methods, Tesfaye *et al*. (Tesfaye *et al*., 2010) proposed a descriptive sensory procedure for the analysis of vinegar. However, the descriptive sensory analysis is depended on the examiner, which requires professional training. In contrast, electrochemical analysis is an alternative approach can be used for food quality evaluation. This technology has been well developed for food colorants (Chen *et al*., 2013; Rouhani, 2009; Thiam *et al*., 2016), additives (Arbeloa *et al*., 2012; Prakash *et al*., 2000; Song *et al*., 2014), toxic compounds determination (Pellegrini *et al*., 2005; Shah *et al*., 2018; Wu *et al*., 2012).

The favor of brewing vinegar is mainly the result of several volatile components such as acids, alcohols, esters, aldehydes, ketones and heterocyclic compounds. Most of these compounds could exhibit electrochemical response at low overpotential. However, the traditional electroanalytical methods cannot be directed used for fermented food analysis due to the complicated composition of the analyte. The recent development of electrochemistry allows the recording of the electroactive profile of plant tissue (Cebrián-Torrejón *et al*., 2017; Doménech-Carbó *et al*., 2017, 2015; Domínguez and Doménech-Carbó, 2015; Martini *et al*., 2015; Ortiz-Miranda *et al*., 2016), food (Chaibun *et al*., 2018) and fungi (Mateo *et al*., 2018). Our previous works demonstrated this method can be used for plant species determination and phylogenetic study (Fu *et al*., 2019, 2018a, 2018b). The electrochemically active substances in the analyte contribute current values at different potentials due to the redox reaction. This electrochemical fingerprint depends on the types and concentrations of electro-active compounds. Therefore, this method has a strong potential value for the identification of the analyte. We would like to extend this methodology for liquid food identification. In this work, electrochemical fingerprints of vinegar were recorded by direct inset the three-electrode system into the sample without adding any electrolyte. White vinegar, rice vinegar and mature vinegar with twelve different vinegar samples were used as real samples. The difference between the fingerprints can be used for vinegar brand determination, which has great potential for filed food authenticity screening.

2 Experimental section

All electrochemical determination processes were carried out using a CHI760 electrochemical workstation. A commercial glassy carbon electrode (GCE), an Ag/AgCl electrode and a Pt electrode were used as the working electrode, reference electrode and counter electrode, respectively. All twelve vinegar were purchased from the local supermarket without any treatment. Table 1 shows the information of all twelve samples.

A differential pulse voltammetry (DPV) scan was used for electrochemistry fingerprint recording. Glassy carbon electrode was firstly polished using alumina slurry after water wash. Then, the three-electrode system was inset into a 5 mL of vinegar. The electrochemical voltammogram recording was conducted at -0.3-1.8 V, with a pulse amplitude of 50 mV, a pulse width of 0.05 s and a pulse period of 0.5 s. After the first scan, the second scan was recorded using the same parameters without pulling out the electrodes.

Fingerprint standardization was carried out for establishing quantitative criteria of recognition (Scampicchio *et al*., 2005), where the ratios between the current and the maximum peak current were obtained at different potentials. PCA and cluster analysis was performed using R based on the recorded electrochemistry fingerprint.

3 Results and discussion

Figure 1 shows the DPV profiles of twelve vinegar recorded using GCE from -0.3 V to 1.8 V. The electrochemical response was successfully recorded when the absence of the additional electrolyte.

Table 1. Information of twelve vinegar samples.

Brand (abbreviation)	Grade	Ingredients	Origin
Chubang-Chencuwang (CB-MV)	Mature vinegar	Water; Sorghum; Barley; Pea; Edible salt; Caramel colour; Sodium benzoate;	Zhongshan, Guangdong
Donghu-Shanxichencu (DH-MV)	Mature vinegar	Water; Sorghum; Barley; Pea; Edible salt; Caramel colour; Sodium benzoate; Bran;	Cavings Taiyuan, Shanxi
Qianhe-Wuguchencu (QH-MV)	Mature vinegar	Water; Sorghum; White granulated sugar; Rice; Edible salt; Corn; Buckwheat; Wheat	Meishan, Sichuan
Shuita-Shuitachencu (ST-MV)	Mature vinegar	Water; Sorghum; Barley; Pea; Edible salt; Caramel colour; Sodium benzoate; Bran	Taiyuan, Shanxi
Chubang-Chunmicu (CC-RV)	Rice vinegar	Water; Rice;	Yangjiang, Guangdong
Haitian-Baimicu (HT-RV)	Rice vinegar	Water; Rice; Edible alcohol; Edible salt	Foshan Guangdong
Hengshun-9°micu (HS-RV)	Rice vinegar	Water; Rice; White granulated sugar	Zhenjiang, Jiangsu
Shuangyu-Baimicu (SY-RV)	Rice vinegar	Water; Rice; Edible salt; Sodium benzoate	Huzhou, Zhejaing
Donghu-Baicu (DH-WV)	White vinegar	Water; Rice; Sorghum; Edible salt; Sodium benzoate	Jinzhong, Shanxi
Jinshansi-Zhenjiangbaicu (JSS-WV)	White vinegar	Water; Rice; White granulated sugar; Edible alcohol; Edible salt; Sodium benzoate	Zhenjiang, Jiangsu
Laohenghe-Baicu (LHH-WV)	White vinegar	Water; Rice; White granulated sugar; Edible salt;	Huzhou, Zhejaing
Zhiwei-Songshengyuan (ZW-WV)	White vinegar	Water; Rice; White granulated sugar; Edible salt; Sodium benzoate	Shaoxin, Zhejiang

It can be ascribed to the presence of acetic acid in the vinegar, which could act as the electrolyte to form an electro-hydraulic circuit.

A series of oxidation peaks were noticed in all DPV profiles, indicating each vinegar contains some of the compounds can be electrochemically oxidized during the scan. According to previous literature, the fermentation process of vinegar production can produce a variety of free amino acids (Kutlán and Molnár-Perl, 2003) and aroma compounds (Natera Marín *et al*., 2002; Tesfaye *et al*., 2004). Among them, several compounds, such as tyrosine (DeFelippis *et al*., 1991), cysteine (Vasjari *et al*., 2005) and vanillin (Deng *et al*., 2015), exhibited district oxidation peak at low overpotential. In addition, some of the ATP-related compounds formed during the fermentation process were also electrochemical active, such as inosine (Liu *et al*., 2006), hypoxanthine (Raj

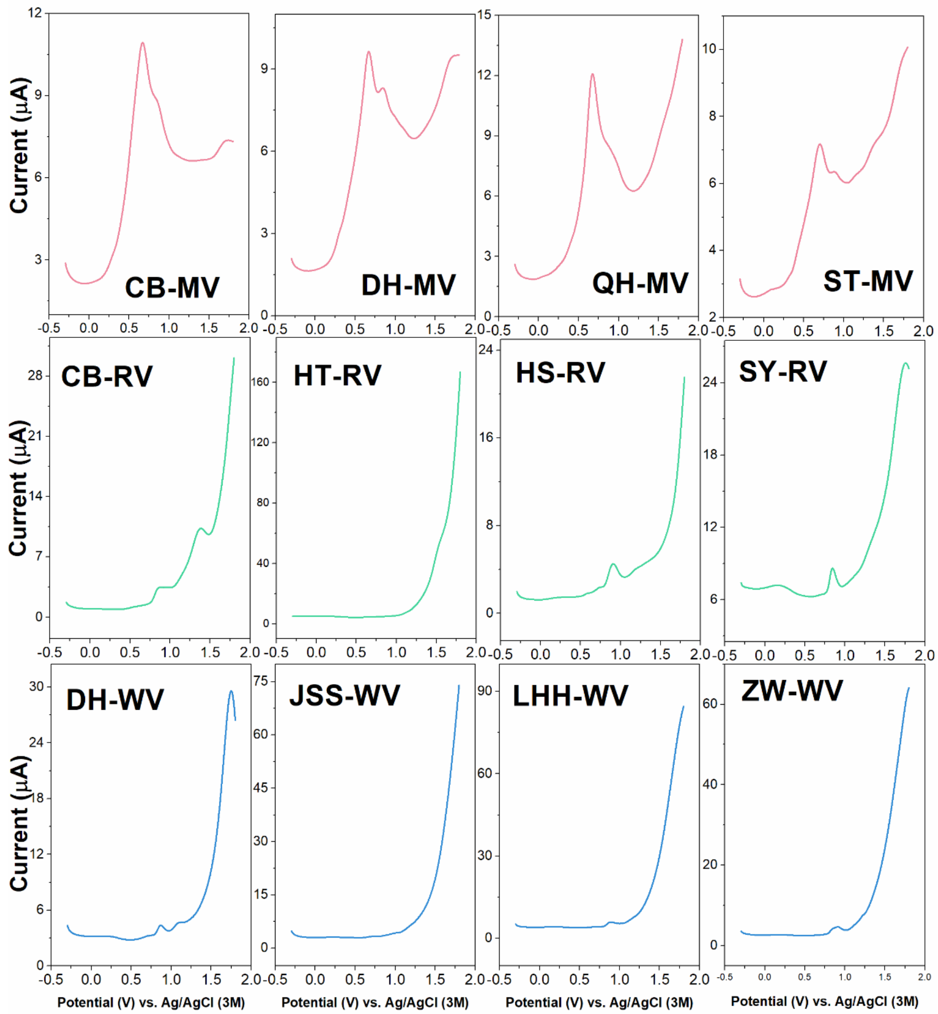

Fig. 1. The first scan of DPV curves of twelve vinegar recorded on a GCE.

and John, 2013), xanthine (Devi *et al*., 2013; Wang and Tong, 2010) and uric acid (Sajid *et al*., 2016; Wang *et al*., 2017).

The current responses of these electro-active compounds result in the electrochemical fingerprint and reflect the composition and ratio between them. Because the similar structure of electro-active compounds may produce signal at similar potential, the identification of each component cannot be achieved at this stage. However, due to the different brewing process and raw material selection (Liu *et al*., 2004), the recorded overall voltammetric pattern showed great potential for vinegar authentic screening.

It is pertinent to note that the DPV curves of all mature vinegar showed a distinct oxidation peak around 0.6 V along with a small peak around 0.8 V. The wealth of electrochemical signals of the mature vinegar indicate more compounds were oxidized compared with that of the rice vinegar and white vinegar. According to the ingredients of the vinegar, the production of mature vinegar uses multiple raw crops including sorghum, barley, pea, wheat, bran and corn, while the white vinegar and rice vinegar usually use the single crop as raw material (López-Cuenca *et al*., 2019). Moreover, a significant longer fermentation process is required for mature vinegar production compared with that of the white vinegar and rice vinegar (Cao *et al*., 2017). Therefore, the mature vinegar contains much higher fermentation products which result in a wealth of electrochemical profile.

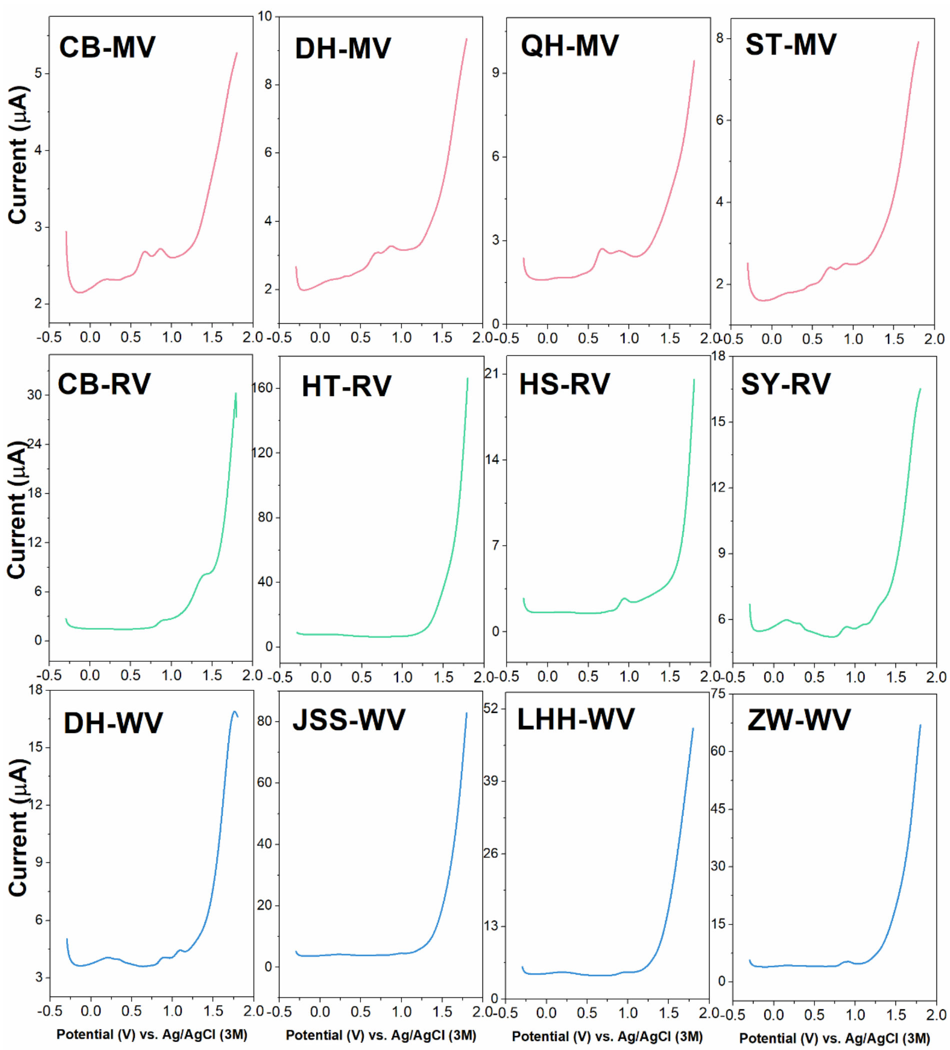

Fig. 2. The second scan of DPV curves of twelve vinegar recorded on a GCE.

In contrast, rice vinegar and white vinegar exhibited much less information during the scan, especially the HT-RV and JSS-WV. The rest three brands of rice vinegar showed a distinct oxidation peak around 1.0 V. The rest three brands of white vinegar also exhibited an oxidation peak around 1.0 V but with low intensity.

The authentic identification of some vinegar, such as ST-MV, CB-RV, can be simply achieved based on observation of DPV profile. However, many other brands cannot be distinguished easily by eye.

Since the oxidized compounds already fouling at the GCE surface, the electrochemical profile of each sample should change significantly. Some additional feature can be revealed. Therefore, we conducted a second scan using DPV without changing the electrode. As shown in Figure 2, the intensity of peaks of all sample declined due to the fouling effect. However, some new features were observed. For example, CB-MV showed a distinct oxidation peak at 0.2 V, while the peak at the similar potential of SY-RV was split into two peaks. Based on these observations recorded the second scans with features recorded in the first scan, we proposed a 2D pattern recognition method for the discrimination of different vinegar (Figure 3). For 2D pattern generation, the normalized current value recorded in the first scan and second scan were used as x and y axis, respectively. These scatter patterns are more easily visualized than the single DPV profile. The brand authentic identification can be conducted by checking the similarity of the converted patterns.

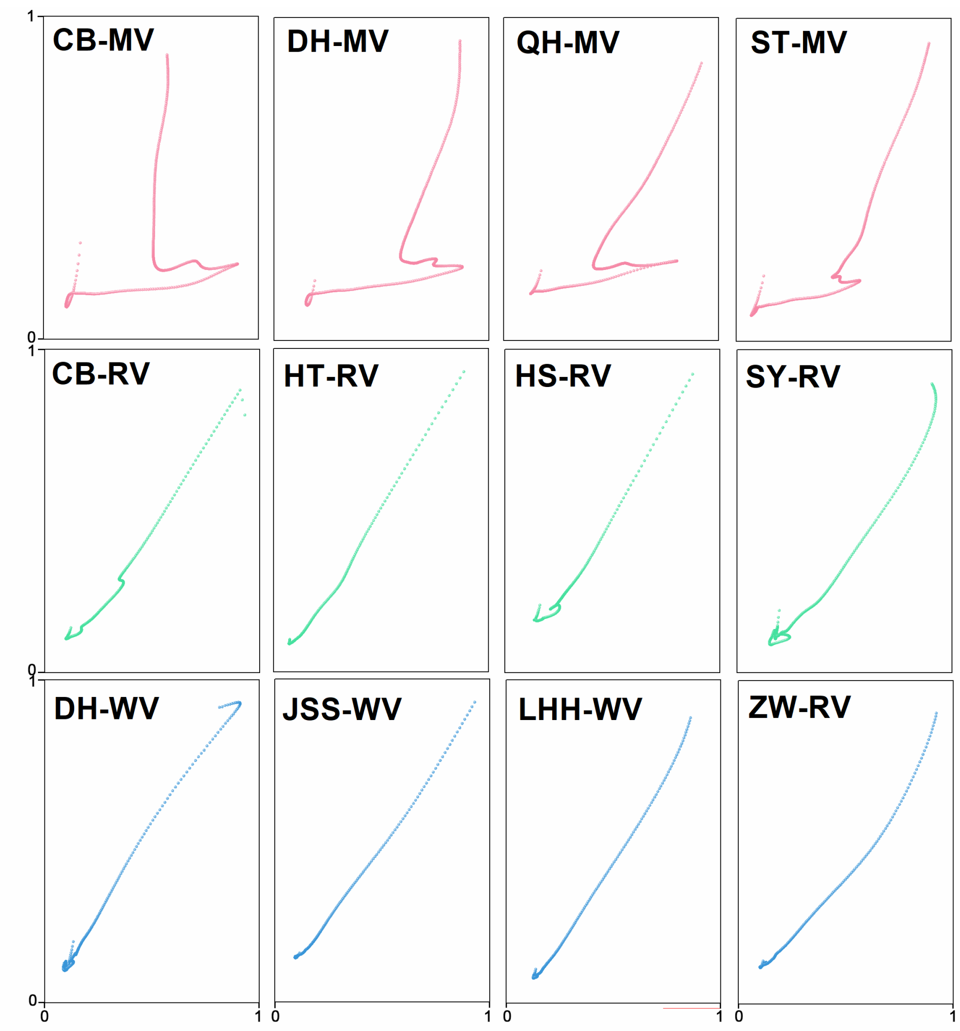

Fig. 3. 2D scatter patterns of twelve vinegar based on normalized currents recorded from first scan vs. normalized currents recorded from second scan.

Then, ten individual samples of each brand have been subjected to the electrochemical fingerprint recording. The principal component analysis (PCA) analysis has been used for evaluating the statistical feasibility of the proposed methodology. As shown in Figure 4, PCA result shows several groupings of the vinegar, which the mature vinegar can be clearly separated from all samples. The white vinegar and rice vinegar have some overlapping probably due to the similar ingredients and short-time fermentation process. The PCA extracted two components that explain approximately 73% of the variation in data.

Hierarchical cluster analysis was then attempted for 120 DPV analysis. As shown in Figure 5, all mature vinegar was in one cluster, indicating they share the common features in the electrochemical fingerprint. In addition, samples of the same brand were clustered together, suggesting the proposed approach can be used for brand identification. In contrast, white vinegar and rice vinegar showed less differentiation, which is confirmed the PCA results. However, the cluster analysis still can be used for brand identification since only less than 6% of samples were clustered into wrong positions. Therefore, we believe the voltammetric data from the vinegar can be used as database and subsequently used for unknown sample identification.

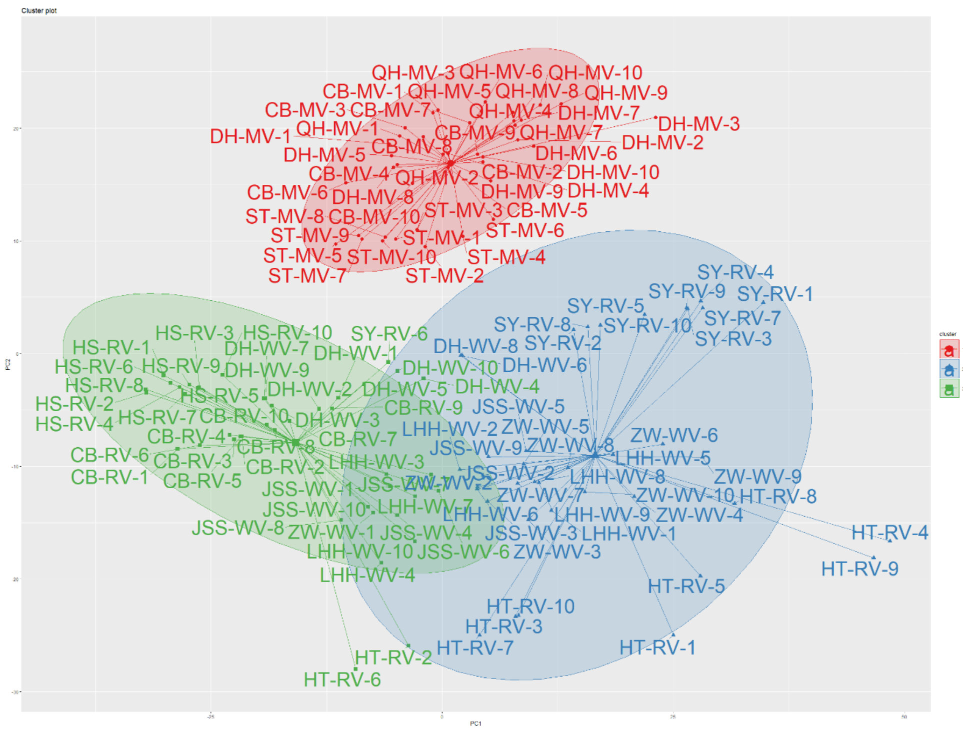

Fig. 4. PCA diagrams of twelve vinegar obtained from normalized currents recorded by voltammetric scans.

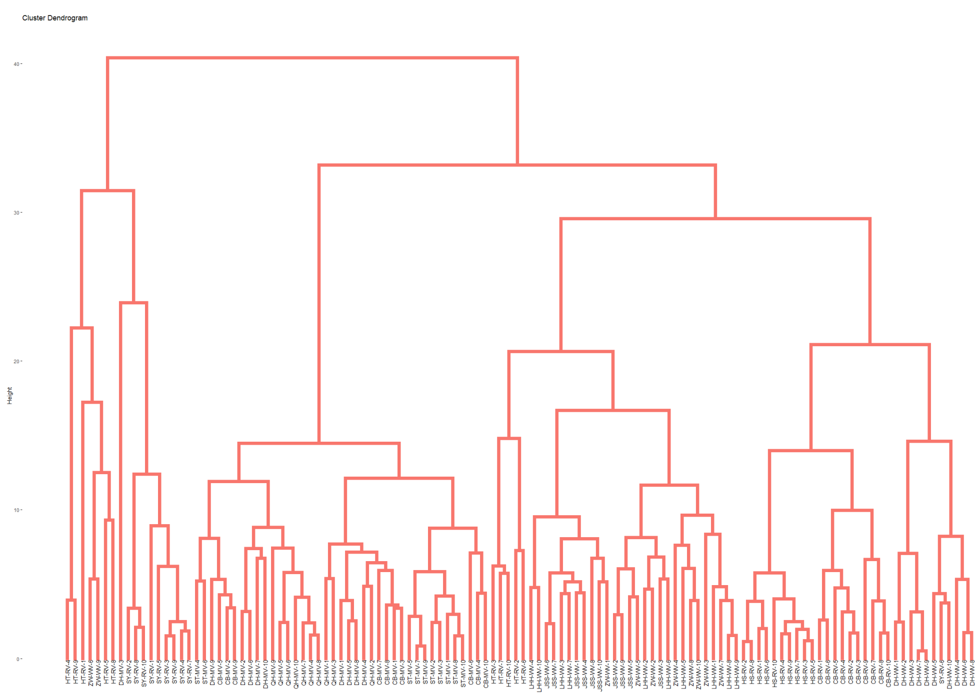

Fig. 5. Clustering analysis twelve vinegar obtained from normalized currents recorded by voltammetric scans.

Conclusion

The electrochemical fingerprint of vinegar can be obtained using commercial glassy carbon electrode due to the presence of the electro-active compounds. Recorded fingerprint varies between the vinegar due to the presence of different content of electro-active compounds produced during the fermentation process. The difference between the electrochemical fingerprint can be used as a barcode for vinegar authentic identification. The DPV profiles can clearly separate the mature vinegar from other types. Due to the high reproducibility of the proposed methodology, the 2D scatter pattern and clustering analysis can be used effectively for vinegar brand identification.

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