



Application of intelligent sensory technology in the authentication of alcoholic beverages

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Abstract

Alcoholic beverages play an important role in social gatherings and the consumption of alcohol drinks keep increasing worldwide in recent years, especially during the COVID-19 pandemic months. The authentication of alcoholic beverage is usually evaluated by trained panels or chromatography analysis. Over the last few decades, intelligent sensory technology (IST) that imitate the human sensory organs have been developed for quality control and authentication of alcoholic beverages. The artificial sensing system consist of arrays of sensors with cross-sensitivity and various pattern recognition methods, which can be used to discriminate or classify the samples based on the detection requirements. Application of IST on wine authenticity have been extensively studied, however, application of IST in authentication of other alcoholic beverages lacks of systemic study. This paper firstly describes the basic mechanism of current IST instruments and then summarizes the applications of IST in alcoholic beverages authenticity assessments, including discrimination of varietal and geographical origins, detection of frauds and adulterations, discrimination of years of aging, distinction of brands and types, aroma analysis, detection of spoilage and off-flavors, and monitoring of the production process. The potential applications and future development of IST in the brewing industry are also discussed.

Keywords: intelligent sensory technology; electronic nose; electronic tongue; alcoholic beverage.

Practical Application: Intelligent sensory technology is an effective and economic tool for real-time quality control of alcoholic beverages.

1 Introduction

Alcoholic beverage plays an important role in social gatherings and alcohol consumption is considered as part of the daily diet in many countries. According to the World Health Organization, the citizens of Czech consumed the most alcohol per person in 2019, with individuals consuming about 14.26 liters of pure alcohol (The Alcohol Industry in Data, 2022). The United States had an annual consumption of 9.97 liters of pure alcohol per person in 2019 (World Health Organization, 2022). As the world continue to adjust to the effects of Covid-19, the sales of alcoholic beverage keep increasing worldwide. The most popular alcoholic beverages around the world are wine (generally 10-15% ethanol by volume), distilled spirit (generally 40-50% ethanol by volume), and beer, lagers and ciders (generally 1.5-8% ethanol by volume) (Figure 1). Global consumption of alcoholic beverages keeps increasing during the past few years.

A growing demand for alcoholic beverages has stimulated fraudulent activities, such as adulteration of geographic origin, false classification of alcohols, and poor-quality products with false labels (Sanaeifar et al., 2017). The falsification harms not only the interests of consumers, but also the reputation of the producers. However, the current instrument-based analyses for quality control

of alcoholic beverages, such as the near infrared spectrum and gas chromatography, needs professional operation that is costly and time-consuming. Sensory evaluation by human panels are usually expensive and it is difficult to establish a corresponding mathematical model (Aleixandre et al., 2018; Los et al., 2021; Cais-Sokolińska et al., 2021). This highlights the shortcomings of alcoholic beverage industry; lacking of rapid, accurate, and economical detection in the market for real-time quality control.

Intelligent sensory technology (IST) is emerging in recent years and set off a new wave for food analysis. IST is to use modern precision instruments to imitate human organs to analyze the sensory characteristics of foods such as color, aroma, taste, and shape (Watson et al., 2021). During the last decades, electronic nose, electronic tongue, and electronic eyes are rapidly developed for alcohol authentication, which shows multiple advantages such as non-destructive, high-speed, good repeatability, reliable detection results, and no complex sample preprocessing process (Watson et al., 2021).

Application of IST on wine authenticity have been extensively studied in a broad of literatures (Rodríguez-Méndez, 2016; Nery & Kubota, 2016; Geană et al., 2020). However, IST investigations

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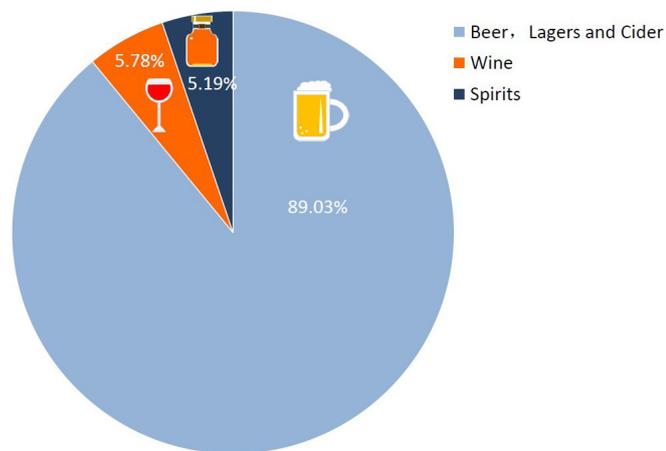


Figure 1. Global consumption of wine, spirits, beer, lagers, and cider in 2021 (The Alcohol Industry in Data, 2022).

for other alcoholic beverages such as beers, distilled spirit, rice wine, and are lacking of systemic study. Thus, this review summarizes the applications of IST on the authentication of a variety of alcoholic beverages, in term of discrimination of varietal and geographical origins, detection of frauds and adulterations, discrimination of years of aging, distinction of brands and types, aroma analysis, detection of spoilage and off-flavors, and monitoring of the production process.

The scientific literature was collected from Web of Science, Scopus, PubMed, Google Scholar, and Research Gate using keywords including “alcoholic beverages”, “intelligent sensory technology”, “electronic nose”, “electronic eyes”, “electronic tongue”, “authentication” and so on. We have reviewed the most relevant progressive studies with an emphasis on the past 5 years. The search results were manually refined for relevance and evaluated to remove multiple and duplicate references.

2 Overview of intelligent sensory technology

IST has expanded exponentially in the past five years for quality control in food industry. Compared to the traditional food analysis technology and sensory evaluation methods, IST has prominent advantages such as: 1) non-invasive detection, 2) can be used for the analysis of toxic compounds or products, 3) faster and more economic (minimize the usage of chemical materials), 4) objective and scientific compared to human sensory panels, and 5) without complicated treatments and well-trained personnel.

2.1 Electronic nose (E-nose)

The concept of E-nose was first introduced in 1982, which is to imitate the structure and mechanism of the mammalian olfactory system (Persaud & Dodd, 1982). The E-nose is composed of an array of electronic chemical sensors with appropriate pattern-recognition system, which can identify individual volatile or complex chemical mixtures that constitute aromas, odors, fragrances, formulations, spills and leaks (Röck et al., 2008). The working principle of E-nose is to simulate the human olfactory

organ through the gas sensor array, then perceive and analyze the odors, and transmit the collected signals information to the analysis software for classification or identification (Figure 2).

One of the advantages of E-nose is no requirement of complex pretreatment for samples. Headspace gas sampling systems usually provide a stable and reproducible sampling. Most of the E-nose devices have two separate chambers, called sample and sensor chamber, as the sampling system (Sanaeifar et al., 2017). The volatiles are conveyed to the sensor chamber along with the gas flow. After reading responses, an inert gas is used to purge both chambers to remove residual odors, in order to prevent the potential cross contaminations (Sanaeifar et al., 2017). In addition, E-nose has a data analysis system: signal pre-processing including baseline manipulation, compression, and normalization, as well as multivariate pattern analysis techniques used for E-noses data post-processing including multiple machine learning models (Sanaeifar et al., 2017). E-noses has a great development in recent years and has been available commercially. To date, E-nose were highly developed and used in diverse industries including chemicals, food and beverage, packaging materials, medical research and so on.

2.2 Electronic tongue (E-tongue)

An E-tongue is a multi-sensor system that is based on an array of sensors/biosensors designs and combined with multivariate statistical data analysis, which is widely used as a rapid and reproducible quantitative and qualitative measurement (Geană et al., 2020). The E-tongue imitates the human taste system, which is mainly composed of three parts: 1) sample preprocessor, which is equivalent to the taste receptor of taste cells in the oral cavity, to convert the features of liquid sample into electronic signals; 2) sensors/biosensors array, which is equivalent to the neural sensory system in the brain, which sensitively captures the signals sent by the preprocessor and transmits it to the data processing system; 3) data processing and pattern recognition system. As shown in Figure 3, the E-tongue is equivalent to the central nervous system in the brain, which uses multivariate pattern analysis models for the interpretation of screening data, including principle component analysis (PCA), linear discriminant analysis (LDA), soft independent modeling of class analogy (SIMCA), partial least squares (PLS) regression and so on (Lvova et al., 2015; Śliwińska et al., 2014). In wine industry, E-tongue systems are often used for the classification of alcohols, detection of falsification, classification of wines by their geographical origin, and discrimination of wines based on specific fingerprints (Rodríguez-Méndez, 2016).

2.3 Electronic eyes (E-eyes)

The E-eye uses image sensors coupled with computer simulation criterion to discriminate/identify the sample color, shape, size, and so on. The measurement is performed with the use of software that creates color spectra with the indication of the size of their presence in the tested sample (Xu, 2019). E-eyes allowing complex sensory information to be processed. Combined with machine learning techniques, E-eyes develop models which learn from a training data set and are capable of fitting complex functions between input and output data,

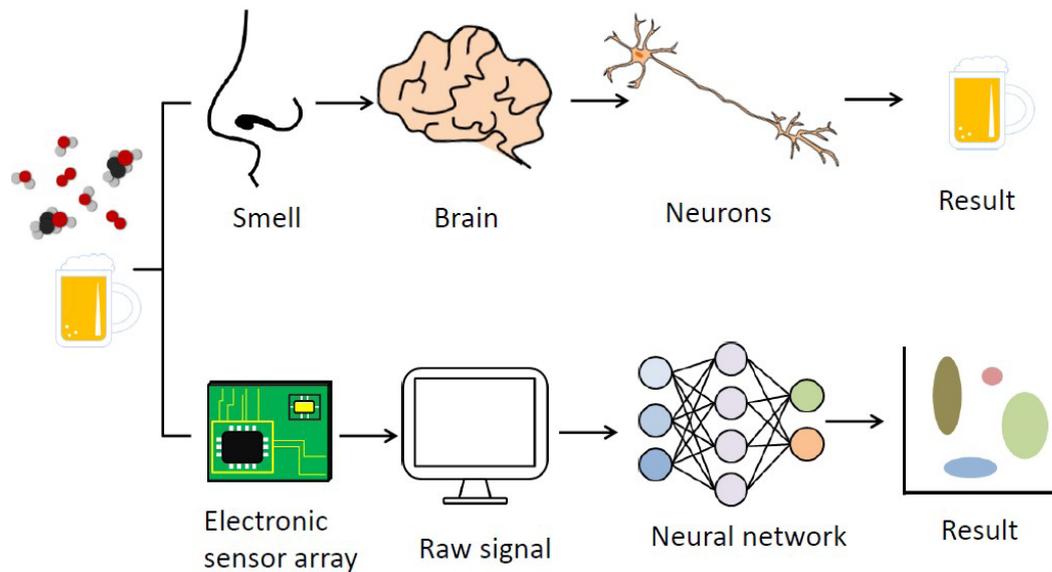


Figure 2. Scheme of the working principle of E-noses.

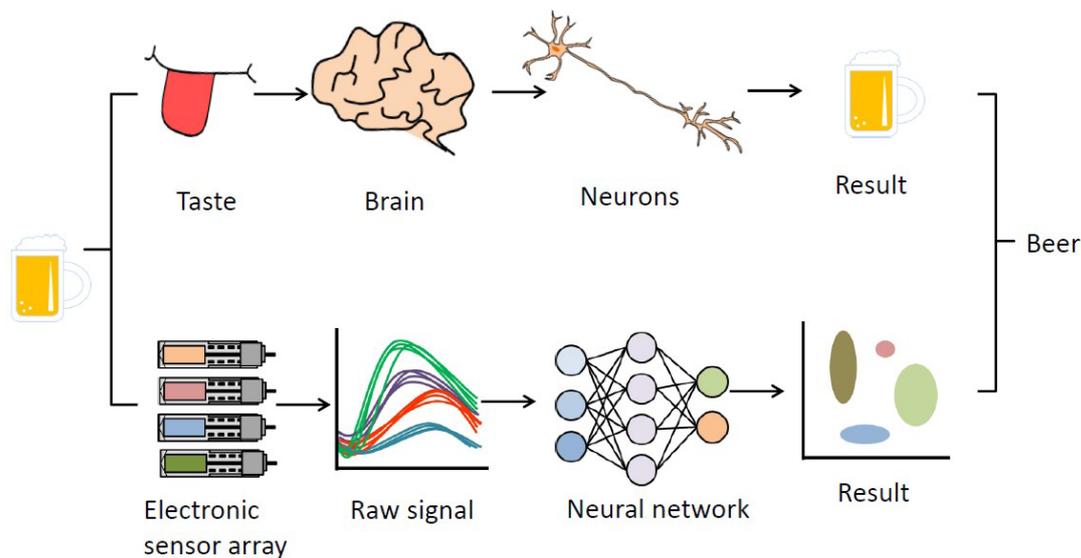


Figure 3. Scheme of the working principle of E-tongues.

which may generate actionable information for manufacturers (Xu, 2019). E-eyes have been used to applied in food industry for characterizing the components that contribute to sensory properties: from ripening to harvest, from raw material to processed food products, and so on.

3 Applications of intelligent sensory techniques for alcoholic beverage authentication

3.1 Discrimination of varietal and geographical origins

Different climatic conditions and geographical environment significantly influence the formation of aromas and chemical compositions of alcoholic beverage during fermentation and aging.

The presence of minor and trace elements during processing under different geographical environment may have certain sensory implications (Song et al., 2018). Therefore, alcoholic beverage produced from different areas show different flavors, especially for wine, beer, and Chinese liquors (baijiu). The identification of the origin of alcoholic products is conducive to the improvement of quality control. The use of IST is of great importance for rapid discrimination of alcoholic beverage origins.

E-nose and E-tongue have been widely used to distinct geographical origins of wines and beers in previous studies (Geană et al., 2020; Martínez-García et al., 2021). In recent five years, the research interest was focused on the development of IST combined with multiple machine learning models to distinct

the wines. Han et al. (2020) used E-nose and E-tongue coupled with extreme learning machine (ELM) to rapidly identify red wines that differed in geographical origins. Principal component analysis (PCA) was utilized for dimensionality reduction and decorrelation of the raw sensors datasets. The fusion models derived from ELM were built with PCA scores of E-nose and E-tongue as the inputs. Results showed that combination of odor (E-nose) and taste sensors (E-tongue) showed superior performance (100% recognition rate) than individual intelligent sensory system.

Followed by the success of wine and beer discrimination, E-nose and E-tongue have been applied in the distinction of origins of various distilled spirits. Miao et al. (2015) used the E-nose installed with olfactory fingerprint analysis system to detect the aroma of rums from different origins including China, Jamaica, Cuba, Guatemala, and Philippines. Through PCA and discriminant analysis, the E-nose could distinguish rum from different origins well. The E-tongue equipped with specific sensor array containing seven IFSET sensors was used to discriminate white wine from different regions in Hungary (Soós et al., 2014). PCA identified initial patterns and linear discriminant analysis (LDA) built models to separate white wine samples based on wine regions and grape cultivars. The method was proven to successfully distinguish white wine with no misclassification error.

As China's national liquor, baijiu dominates China's domestic spirits market, which contributed to 96% of the total sales value (Ma et al., 2019). The volatiles of baijiu are critical indicators in tracing the geographical origin and the IST methods that are capable of predicting the geographical region of the different types of baijiu have been constructed. Peng et al. (2015) applied E-nose technique and chemometrics analysis as a rapid tool for the discrimination of baijiu from different geographical origins. Through PCA and DFA analysis, the E-nose technique combined with chemometrics methods could be used as fingerprinting techniques to distinct baijiu and enable its authentication. The E-tongue based on three nanocomposites modified electrodes was applied for the identification of rice wines of different geographical origins including Zhenjiang, Qingdao, Jiaying, Taizhou, and Shaoxing (Wang et al., 2019). The modified electrodes showed high sensitivity to the indicators of geographical origins for rice wine, such as tyrosine and gallic acid. Among PCA, LDA, and locality preserving projection (LPP) analysis, LPP was best performed in the classification of rice wines.

3.2 Detection of frauds and adulterations

Motivated by the high price of wine and liquor, there were quite a few market cheating scandals occurring, such as adulteration, counterfeit, and fraudulent flavor (Alexander et al., 2018; Geană et al., 2020). Alcohol adulteration is commonly occurred by mixture of methanol, ethanol as well as with other alcohols of the same color, which has damaged the reputation of alcohol market and the benefit of consumers. To avoid adulteration and market cheating, a rapid, reliable, and accurate detection technology for authentication of alcoholic beverage is essentially required for the prevention of illegal adulteration.

Electroanalytical system equipped with various sensors and coupled with multiple pattern recognition methods, such as E-nose and E-tongues, have been widely applied in wine authentication (Rodríguez-Méndez, 2016). Different electroactive materials, electrochemical sensors, electrodes types, and functionalization sensitive materials have been explored in the previous studies (Geană et al., 2020). In recent years, electroanalytical techniques are widely used in frauds and adulterations of Chinese liquors (baijiu). Qi et al. (2017) applied a portable E-nose equipment or rapid real-fake recognition of Chinese liquors. The E-nose system consisted of evaporation/sampling section, a reaction module, a control/data acquisition and analysis module, and a power module. The optimization of parameter through a one-class support vector machine classifier largely improved the overall recognition accuracy of the modified E-nose system. Ma et al. (2019) used a portable E-nose instrument to quickly detect the adulteration of Chinese liquor. The sensor array was used to obtain the "fingerprint data" of liquor first, and then the characteristic information was extracted by discrete wavelet transformation. PCA was used to achieve quantitative discrimination of liquor samples of different purity and therefore to discriminate the fake liquors samples.

3.3 Discrimination of years of aging

The economic value of a majority of alcoholic beverage is highly associated with the years of aging (Sun et al., 2017; Cao et al., 2021). During aging process, a series of complex physicochemical reactions occurred, such as oxidation, esterification and hydrolysis, which increased the abundance and variety of volatiles in the alcoholic drinks (Apetrei et al., 2012; Cao et al., 2021). Therefore, the years of aging decides the quality and flavor of alcoholic beverage. Some dishonest producers counterfeit their younger products as several years aged liquors to get a higher selling price (Cao et al., 2021). Thus, the discrimination of the age of alcohols is essential to protect the benefit of consumers as well as the reputation of the producers.

E-nose, E-tongue, and a combination of them were used for the discrimination and classification of wines according to the vintage year, recognize the method of aging, and monitor the aging process (Rodríguez-Méndez, 2016; Geană et al., 2020). Voltammetric sensors, chemically modified with electrocatalytic materials, have been widely used to monitor the changes in wine during aging in different types of barrels and bottles (Śliwińska et al., 2014; Geană et al., 2020).

Chinese liquor with different ages were well-discriminated by PCA, LDA and analysis of variance (ANOVA) based on E-nose signals (Zhou et al., 2020; Cao et al., 2021). E-nose coupled with LDA was also proven to be an effective and economic way for discrimination of Luzhou-flavor liquor after 2, 3, and 5 years of aging, and the overall aroma characteristics of the liquors stored for different years were validated by gas chromatography/time-of-flight mass spectrometry (Cao et al., 2021). In addition, E-nose coupled with PCA, LDA and ANOVA successfully discriminated the strong-sauce-flavor aged Chinese liquor (Hubei, China) with an aging period of 12, 15, and 20 years (Zhou et al., 2020).

3.4 Distinction of brands and types

The brand of alcoholic beverage does not only reflect a single coherent entity, it more refers to perceived quality, brand awareness, consumers' loyalty and brand associations. E-nose and E-tongue technology have commonly been used to discriminate the different brands of wine and beer with the same content of alcohols and vintage years (Sanaeifar et al., 2017). Recently, Paknahad et al. (2017) applied a metal oxide gas sensor with an open digital microfluidic (DMF) system in rapid recognition of different types of wines ($n=7$). This system successfully provided selective detection of the aromas of a wine droplet manipulated on a DMF platform and two features are extracted from the transient response and mapped into a 2-D plot to show the discrimination of different wines. In another study, the E-nose with transient features inclusive and dimensionality reduction by PCA has been applied in beer brand classification. A simple-but-powerful learning vector quantization algorithm was used to derive the rate of classification accuracy, which showed 100% of classification accuracy when including transient features, compared to the traditional method with 61% accuracy (Nimsuk, 2019).

In case of distilled spirit, Wu et al. (2019) used E-nose based on a novel feature extraction algorithm, called fuzzy discriminant principal component analysis (FDPCA), to identify different types of Chinese liquors, including Maotai, Fenjiu, Kouzijiiao, Haizhilan, Yingjiagong and Gujinggong. The accuracy of the E-nose system coupled with FDPCA showed good performance (98.4%) in classification of Chinese liquors (Wu et al., 2019). In order to revise the unreasonable structure of current E-nose chamber and apply it in handheld E-nose system, Wang et al. (2021) designed seven schemes of E-nose bionic chamber for the discrimination of Chinese liquors. Inspired by the structural characteristics of mammalian nasal cavity, the effects of chamber shape and internal structure on airflow uniformity, flow velocity, air pressure and residual gas stimulated by fluid dynamics were compared. The bionic chamber with the best performance was selected and installed in a self-made handheld E-nose system, which showed high recognition rate in discrimination of Chinese liquors including Fen (42% Vol), Guo Jiao (52% Vol), Jiannan Chun (52% Vol), Maotai (53% Vol), Wuliangye (52% Vol), and Xifeng (52% Vol).

3.5 Aroma analysis

Each alcoholic beverage has its own unique flavor characteristics. The use of IST technology to study the flavor composition of alcoholic beverage, especially the changes during production process and storage, is always a research hotspot in the alcohol industry. IST technology is able to analyze aroma profiles by registering signals produced by the mixture of gases and then comparing the pattern of responses produced by different samples.

IST technology have been widely applied to the identification of wine aromas. E-nose has been used to recognize typical aromas in both red wine and white wine grouped into different families (floral, fruity, microbiological, herbaceous, and chemical) (Liu et al., 2018). Recently, E-tongue has been used in the evaluation of the sensory perception (color, aroma,

flavor, mouthfeel, taste, and aftertaste) of ciders (Brown Snout) produced from machine-harvested and hand-harvested fruit in 2014 and 2015 (Alexander et al., 2018). The E-tongue showed a response to metallic and sour taste that was more associated with the machine-harvested samples while a response to sweet and umami taste that was more associated with the hand-harvested samples. Zhu et al. (2016) used E-nose combined with PCA and DFA analysis to show that a high pressure treatment at 300MPa resulted in significant ($p < 0.05$) changes in aroma profiles of Chinese "Junchang" liquor. An increase in total ester content and a decrease in total acid content were observed for all treated samples ($p < 0.05$), which was verified by gas chromatography analysis. Sensory evaluation results confirmed that the favorable changes in flavor were induced by high pressure treatment, which may accelerate the aging process of traditional liquor (Zhu et al., 2016).

3.6 Detection of spoilage and off-flavors

The presence of unexpected off-flavors formed during fermentation or storage is a great concern for alcoholic beverages, which largely impact the quality and consumer acceptance of the products. The early detection of these off-flavors is crucial to undertake remedial actions that can correct the fault. A portable E-nose is usually employed as a real-time detection tool for quality control of alcoholic beverages.

Beers and wines were doped with off-flavor compounds such as acetic acid ("sour" attribute), acetaldehyde ("tart" attribute), sulfur compounds ("rotten egg, sewage and rubber" attribute), hexan-1-ol ("herbaceous" attribute), ethyl acetate ("oxide" attribute), 4-ethyl phenol ("leather/stable" attribute), 2,4,6-trichloroanisole ("musty/cork" attribute), oct-1-en-3-ol ("mushroom" attribute) and diacetyl, 2,3 butanedione ("butter" attribute) (Macias et al., 2012; Rodríguez-Méndez, 2016; Sanaeifar et al., 2017). Gamboa et al. (2019) used a portable and compact self-developed E-nose, based on thin film semiconductor (SnO_2) sensors and trained with an approach of deep multilayer perceptron (MLP) neural network, was served as an effective real-time tool for monitoring acetic acid contents in wines. Through a rising-window that focused on raw data processing to find an early portion of the sensor signals with the best recognition performance, the developed E-nose system could classify the wine spoilage in 2.7 s after the gas injection point, which was 63 times faster than the conventional approach (Gamboa et al., 2019).

Spoilage due to the off-flavors can also be detected by E-tongues. E-tongue has been used to monitor the levels of acetic acid in various wines (Rodríguez-Méndez, 2016). The potentiometric E-tongue system has been developed to distinguish wine samples on the base of permitted levels of fault compounds content, which may serve as a detection technology for brand uniformity control of monoculture Apulian red wines (Lvova et al., 2018). The sensor array was composed of 8 porphyrin coatings produced by electrochemical polymerization, which was employed for discrimination and quantitative detection of wine defect compounds (off-flavors), including 3-(methylthio)-propanol, isoamyl alcohol fusel oil, benzaldehyde, and acetic acid (marker of vinegar formation). E-tongue coupled with PLS permitted a discrimination precision

of at least 70% of tested wines in respect to the brand affiliation. Satisfactory PLS predictions were obtained in real wine samples with $R^2 = 0.989$ for isoamyl alcohol and $R^2 = 0.732$ for acetic acid (Lvova et al., 2018).

3.7 Monitor of the production process

The main production process of alcoholic beverages includes fermentation, aging, and bottling. Various elements can be introduced into alcoholic beverages during the processing, which can affect the taste, smell, and color. The E-nose and E-tongue are reported to be able to assess the fermentation process, detection of faults, traceability, as well as indicate inappropriate storage conditions of alcoholic beverages in an economic and user-friendly manner (Nery & Kubota, 2016; Sanaeifar et al., 2017).

Artificial intelligence integration with low-cost sensor networks in the form of E-nose/ E-tongue system has been applied to the quality control of wines at different stages of production, in order to improve the production process (Sanaeifar et al., 2017; Watson et al., 2021). E-nose coupled with machine learning has showed a high accuracy in the identification (97%) of fermentation type of beers, physicochemical, and colorimetry of beers, which provided a rapid detection for quality control of beers (Gonzalez Viejo & Fuentes, 2020). Martínez-García et al. (2021) used E-nose to evaluate the effect of yeast inoculation format (free cells and yeast biocapsules), temperature (10 and 14 °C), and aging time (15 and 24 months) factors on the composition of sparkling wines. The discriminant models based on E-nose dataset enable a 100% correct classification of samples, in relation with aging time and format factors and the 83% for temperature factor, which provided a real-time detection for industry to observe the influence of processing conditions on the final quality of sparkling wines (Martínez-García et al., 2021).

On the other hand, IST technology was used to secure the high-quality of alcoholic beverages products in the early stage of brewing. Integration of near-infrared spectroscopy (NIR) and E-nose using machine learning modelling was applied in rapid detection of beer faults (Gonzalez Viejo et al., 2021). Aroma profiles from 18 commercial beers and a control sample were used as targets for classification of machine learning modelling and 6 different models were developed. Among them, two models (I and II) showed high classification rendering precisions (95.6% and 95.3%) in classification of the beers into control, low and high levels of faults. The developed intelligent system showed a promising and economic application in robotic pourers, especially for large brewing companies (Gonzalez Viejo et al., 2021). In addition, a combination of E-nose and E-tongue coupled with PCA and cluster analysis methods was developed to monitor the quality of jujube wine (Tang et al., 2020). Through multivariate analysis of variance, significant differences ($P < 0.001$) were observed among jujube wine samples during the three fermentation stages: early stage (0-24 hours), middle stage (36-132 hours), and late stage (144-240 hours). Redundancy analysis demonstrated that the primary quality variation of jujube wine was occurred in the middle stage of fermentation (Tang et al., 2020).

4 Conclusion

This paper presents a comprehensive literature review addressing the authenticity determination of a variety of alcoholic beverages through intelligent sensory technology. Based on different electrochemical techniques coupled with appropriate pattern recognition methods, IST have been successfully applied in quality assessments of alcoholic beverages, in terms of variety and geographical origins, monitoring production processes, detection of frauds and adulterations, discrimination of years of aging, distinction of brands and types, aroma analysis, detection of spoilage and off-flavors, and monitoring of the production process. However, the E-noses and E-tongues instruments still need improvement, especially the development of high sensitivity and selectivity bioelectronics sensor arrays aimed at improving accuracy and reliability of the analysis. Future studies may focus on developing the practical methodologies/protocols for the authentication of different type of alcoholic beverages, such as the guidance for selection of appropriate sensors coupled with pattern recognition methods. Furthermore, to establish reliable statistical models, it is important to process and collect a large number of data covering all type of alcoholic beverages from all possible sources of variability including different brands, origins, vintage years, and storage time. The interest in the IST technology in authentication of alcoholic beverages will be still growing, portable intelligent device equipped with clear protocols will be a promising analytical practice in brewing industry.

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