Food Chemistry: X 18 (2023) 100718

Contents lists available at ScienceDirect

Food Chemistry: X

journal homepage: www.sciencedirect.com/journal/food-chemistry-x

# Monitoring the major taste components during black tea fermentation using multielement fusion information in decision level

Ting An [a,b,c], Zheli Wang [c], Guanglin Li [a], Shuxiang Fan [c], Wenqian Huang [c], Dandan Duan [d], Chunjiang Zhao [a,d,*], Xi Tian [c,*], Chunwang Dong [b,*]

[a] College of Engineering and Technology, Southwest University, Chongqing 400715, China
[b] Tea Research Institute, Shandong Academy of Agricultural Sciences, Jinan 250033, China
[c] Intelligent Equipment Research Center, Beijing Academy of Agriculture and Forestry Sciences, Beijing 100097, China
[d] Information Technology Research Center, Beijing Academy of Agriculture and Forestry Sciences, Beijing 100097, China

ARTICLE INFO

*Keywords:*
Black tea
Hyperspectral imaging
Electrical properties
Data fusion
Stacking combination strategy

ABSTRACT

Hitherto, the intelligent detection of black tea fermentation quality is still a thought-provoking problem because of one-side sample information and poor model performance. This study proposed a novel method for the prediction of major chemical components including total catechins, soluble sugar and caffeine using hyperspectral imaging technology and electrical properties. The multielement fusion information were used to establish quantitative prediction models. The performance of model using multielement fusion information was better than that of model using single information. Subsequently, the stacking combination model using fusion data combined with feature selection algorithms for evaluating the fermentation quality of black tea. Our proposed strategy achieved better performance than classical linear and nonlinear algorithms, with the correlation coefficient of the prediction set ($R_p$) for total catechins, soluble sugar and caffeine being 0.9978, 0.9973 and 0.9560, respectively. The results demonstrated that our proposed strategy could effectively evaluate the fermentation quality of black tea.

## 1. Introduction

Black tea has become one of the most popular beverages in the world, thanks to its attractive flavor and health benefits (An et al., 2022; Qu et al., 2020). Compared with these unique advantages, the complicated production technologies of black tea are not familiar to consumers. Importantly, the fermentation directly affects the quality of finished black tea, such as the taste, color, aroma and physical characteristics, because it produces considerable chemical components that determine the quality of black tea. From a biochemical perspective, catechin exhibit a series of oxidation reduction reactions under the catalytic action of enzymes (peroxidase, polyphenol oxidase and other enzymes) to form water-soluble pigments and volatile aromatic substances, leading to a considerable change in the aroma and color of tea leaves. Meanwhile, ester catechins show a strong bitterness, while non-ester catechins show a slightly sweet aftertaste with some weaker bitterness (Dong et al., 2021). Remarkably, the soluble sugar presents sweet and mellow taste and the caffeine is considered as an important contributor to the bitterness of finished tea (Chen et al., 2018). Obviously, these components directly determine the taste and quality of finished tea. These components need to be detected by professional technicians in the laboratory. Some obvious shortcomings, such as destructive, expensive and time-consuming, determine that the existing detection technology cannot adapt to actual production. A rapid, convenient and nondestructive detection method of major fermentation components needs to be developed.

In recent years, the quality of agricultural products could be effectively evaluated by modern nondestructive detection technology, especially in black tea processing (An, Li et al., 2022; Fan et al., 2022; Zarezadeh & Aboonajmi, 2023; Yu et al., 2022). As for black tea fermentation, such technologies mainly contain near infrared spectroscopy (NIRS) (Dong, Li et al., 2018), computer vision system (CVS) (Dong, Li et al., 2018) and electrical properties (Zhu et al., 2019), which have been applied to evaluate the fermentation quality of black tea. The

* Corresponding authors at: College of Engineering and Technology, Southwest University, Chongqing, 400715, China (Chunjiang Zhao). Intelligent Equipment Research Center, Beijing Academy of Agriculture and Forestry Sciences, Beijing 100097, China (Xi Tian). Tea research Institute, Shandong Academy of Agricultural Sciences, Jinan 250033, China (Chunwang Dong).

*E-mail addresses:* zhaocj@nercita.org.cn (C. Zhao), tianx2019@sina.com (X. Tian), dongchunwang@163.com (C. Dong).

https://doi.org/10.1016/j.fochx.2023.100718
Received 21 March 2023; Received in revised form 14 May 2023; Accepted 18 May 2023
Available online 22 May 2023

above technology could only obtain one-sided information of tea samples from one aspect, because of the characteristics of each instrument. For instance, the CVS could obtain the color and texture features, which are closely related to the vision information of the sample surface. The NIRS and electrical properties detect the major chemical components corresponding to the taste of finished tea. Remarkably, the NIRS could evaluate the taste information of fermentation samples based on the overtone of key groups at some characteristic bands. The electrical properties could predict the non-volatile components according to the changes in cell architecture and constituents. However, these technologies also exhibited some limitations. For instance, both the infrared spectroscopy and computer vision technology show weak penetration ability, which could not detect deep sample information. In contrast, the electrical properties technology shows stronger penetration ability and it can obtain the feature information of the whole samples based on the changes in cell architecture, but its detection indicators are limited. These technologies have acquired relatively independent information and fail to collect the comprehensive features of tea samples from different aspects. Hence, the fusion technology of different effective information is necessary to be developed.

The fusion of multielement information technologies provide a novel strategy to evaluate the quality of tea samples. According to previous studies, the quality grade and fermentation degree of black tea were accurately evaluated based on the fusion information of infrared spectra and visual image (Jin et al., 2020; Li et al., 2021). Jin et al. (Jin et al., 2021) successfully predicted theaflavin (TFs) during black tea fermentation by merging Micro-NIRS and CVS information. Unfortunately, other theaflavins monomers including theaflavin-3-gallate (TF-3-G), theaflavin-3′-gallate (TF-3′-G), and theaflavin-3-3′-gallate (TFDG) have not been accurately predicted, but the prediction performance of these models has a certain improvement compared with the single data model. Although these studies carefully considered the external (color and texture) and internal features (the overtone of key groups) of tea samples, the information obtained by NIRS technology has a strong limitation because of the small collection range of the instrument. The spectral information could not perfectly correspond to the visual image information because the obtained sample range using CVS is larger than the obtained sample range based on NIRS. Our previous study effectively overcame this problem and successfully evaluated the fermentation degree of black tea by combining the sample hyperspectral imaging and colorimetric sensing array to mimic human sensory information (An, Huang et al., 2022). However, hyperspectral imaging technology shows weak penetration ability, which could not detect deep sample information. In this study, the electrical properties information and hyperspectral imaging information were fused to obtain comprehensive information of accumulated tea samples (color, texture, taste and deep sample information). The collected information using our proposed strategy is suitable for the needs of large-scale processing in actual production. Importantly, there are no studies that monitor the major chemical components corresponding to the taste using the fusion information of hyperspectral and electrical properties during tea processing, especially in black tea fermentation. In addition, most data fusion models are established using the information from data level (low-level fusion) and feature level (middle-level fusion), and there are no researches that establish the stacking combination model using the fusion data in decision level.

Black tea fermentation contains many complicated physical and chemical reactions, which change the taste and appearance of tea samples. Hence, we aimed at integrating the hyperspectral and electrical properties information and using stacking combination strategy to more effectively evaluate the fermentation quality of black tea. The specific steps of our study were as follows: (1) To collect hyperspectral and electrical properties data at different fermentation times. (2) To acquire the spectral and image information based on obtained hyperspectral image. (3) To establish the quantitative prediction model of major chemical components using single information. (4) To establish the quantitative prediction model of major chemical components using the fusion information and stacking combination strategy.

## 2. Materials and methods

### 2.1. Samples

In this study, 50 kg fresh leaves of 'Jiukeng' variety with one bud one leaf were collected in Hangzhou on May 5, 2022 and these fresh tea leaves were processed according to the manufacturing crafts of black tea (GB/T 35810–2018). After the withering and rolling, the tea leaves were placed in a fermentation room with 30℃ and 90% for the temperature and humidity, respectively. In order to obtain more comprehensive fermentation information, the fermentation time was extended to 7 h. During fermentation experiment, 2 kg samples were collected from fermentation room every hour using five-point sampling method and they were divided into 20 parallel samples (approximately 100 g per sample) for the information acquisition of hyperspectral image first and then the electrical parameters. After obtaining digital information, these samples were placed in liquid nitrogen and then freeze-dried for determination of chemical components regarding the taste of black tea. Finally, 160 samples were obtained in this experiment. The fermentation experiment was illustrated in S-Fig. 1.

### 2.2. Hyperspectral image acquisition

The hyperspectral imaging system (400–1000 nm) was the same as described in our previous study (An, Huang et al., 2022). For each pixel, the spectral curve contained 557 bands. To avoid baseline drift, the equipment turned on for 30 min before the fermentation experiment. After repeated adjustment, some significant parameters, such as exposure time and movement speed of 4.2 ms and 2.8 mm/s respectively, were determined. During fermentation experiment, the collected 20 parallel samples every hour were used to obtain hyperspectral images respectively. Hence, 160 hyperspectral images of fermentation samples were collected. Subsequently, these obtained hyperspectral images were calibrated based on the formula, which was described in our supplementary materials. The hyperspectral image acquisition of fermentation samples was displayed in S-Fig. 2. Then, these collected samples were used to obtain electrical parameters.

### 2.3. Electrical properties information acquisition

Electrical properties test system including a TH2829C LCR bridge meter (Changzhou Tonghui Electronic Co. Ltd., Changzhou, Jiangsu, China), a self-built rectangular wooden box with length, width and height of 0.2 m, 0.1 m and 0.05 m respectively, a pair of red copper electrode (0.1 m, 0.05 m and 0.0008 m, Taizhou New Material Co., Ltd., Taizhou, Jiangsu, China) and a computer, was used to obtain electrical parameters of fermentation samples. The testing frequency range and the resolution were 20 Hz-1 MHz and 1 mHz, respectively. Before testing the electrical properties of tea samples, the system turned on for 30 min. In order to comprehensively acquire the characteristic frequency information of fermentation samples, 65 effective frequencies including 0.02, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.15, 0.2, 0.25, 0.3, 0.35, 0.4, 0.45, 0.5, 0.55, 0.6, 0.65, 0.7, 0.75, 0.8, 0.85, 0.9, 0.95, 1, 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 100, 150, 200, 250, 300, 350, 400, 450, 500, 550, 600, 650, 700, 750, 800, 850, 900, 950, 1000 kHz were measured. For each frequency, 7 electrical parameters, such as parallel equivalent inductance (Lp), parallel equivalent resistance (Rp), loss factor (D), quality factor (Q), complex impedance (Z), phase loss angle (θ) and reactance (X), were collected. Subsequently, each sequential fermentation sample (approximately 100 g) was measured by electrical properties test system to obtain electrical parameters at different test frequencies. Thus, the electrical parameters of 160 fermentation samples at different frequencies (a 160*455(65*7)

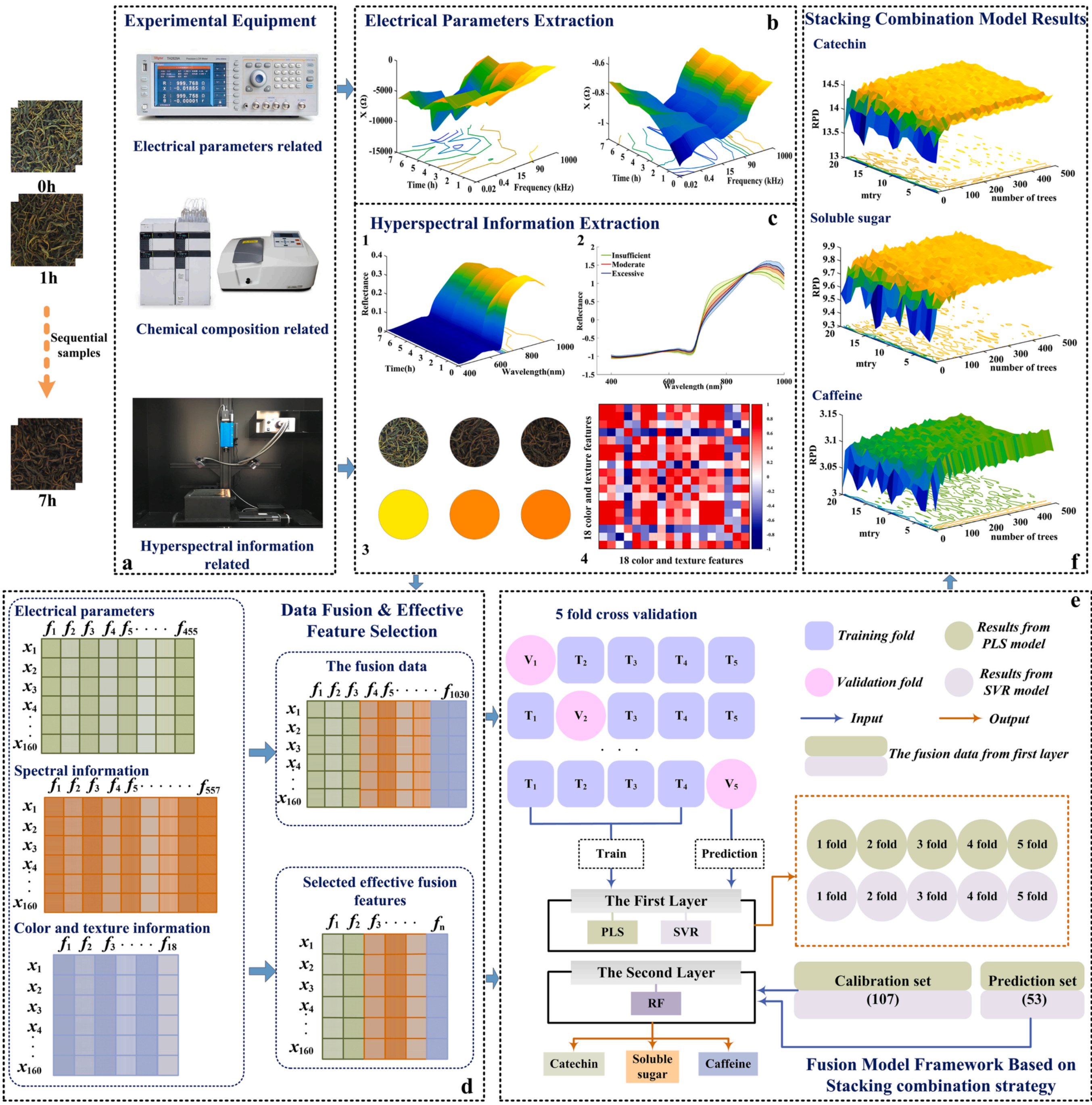


**Fig. 1.** The flow diagram of fermentation experiment and fusion model framework using stacking combination strategy. (a) indicates the required equipment for fermentation experiment. (b) shows representative electrical parameters raw X (left figure) and preprocessed X (right figure). (c) represents the spectra and image data extracted from sample hyperspectral images. (d) shows the data fusion and effective features selection strategy. (e) represents the fusion model framework based on stacking combination strategy. (f) means the predictive results of stacking combination model for fusion-BOSS.

matrix) were obtained and these data were subjected to Z-score for processing to improve the comparability of different variables. The electrical properties information acquisition of these sequential samples was shown in S-Fig. 2.

### 2.4. Determination of major chemical components

After collecting the digital information (hyperspectral images and electrical parameters) of sequential fermentation samples, these samples were freeze-dried for determination of chemical components, such as total catechins, soluble sugar and caffeine. Catechins were determined based on the determination of tea polyphenols and catechins content in tea (GB/T8313-2008). The soluble sugar was determined according to the determination of soluble sugar in vegetables and their products-Copper reduction iodometry (NY/T1278-2007). The caffeine was measured with reference to the determination of the Caffeine (GB/T8312-2013).

### 2.5. Data processing

#### 2.5.1. Hyperspectral image processing

Hyperspectral image of fermentation samples contains a large

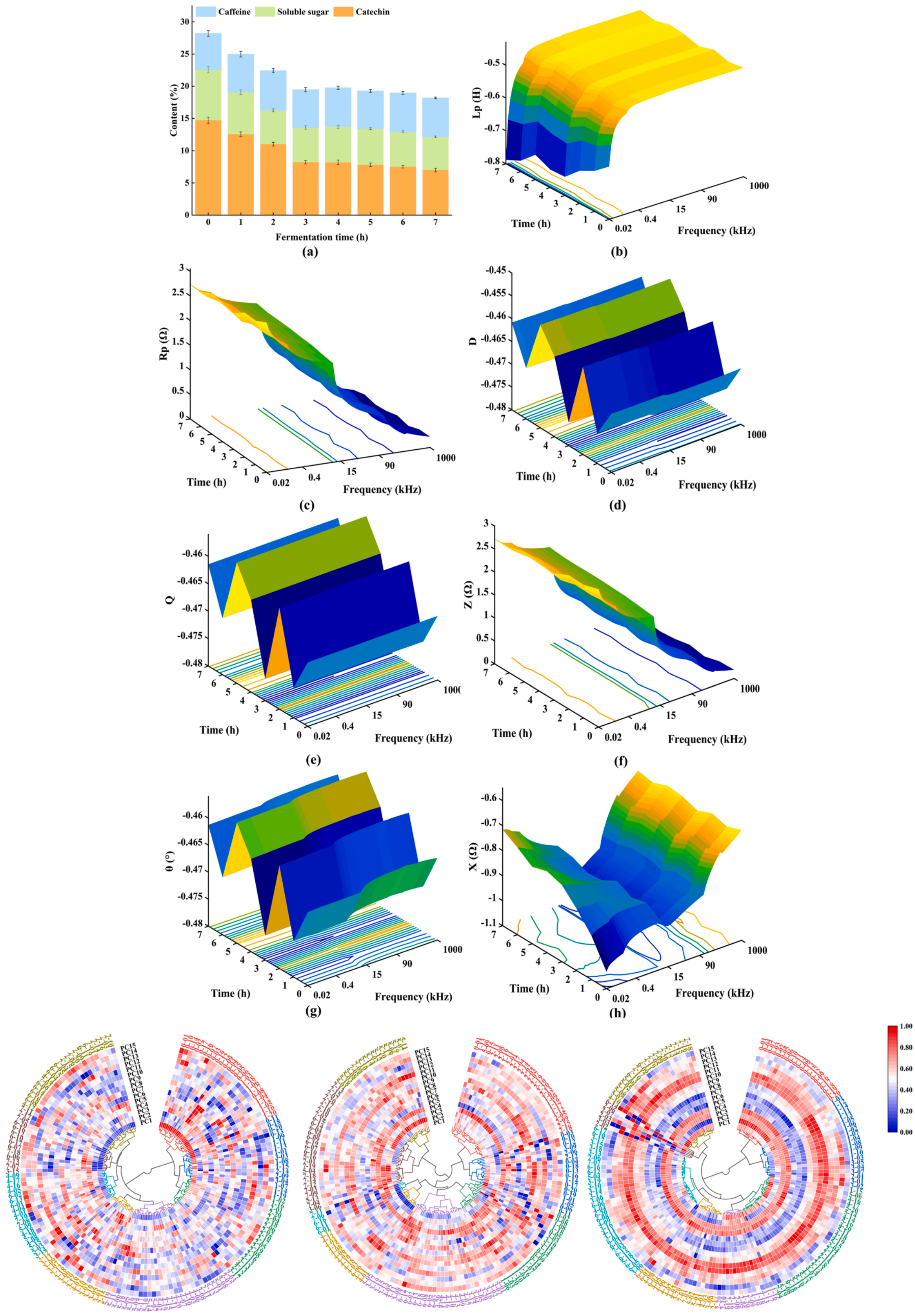


**Fig. 2.** (a) Stacking histogram of major chemical components. The regular patterns of electrical parameters after Z-scores processing with the increasing of fermentation time and test frequency: (b) Lp, (c) Rp, (d) D, (e) Q, (f) Z, (g) θ, (h) X. Hierarchical clustering heatmap based on (i) spectral, (j) image and (k) electrical properties information of the first 15 PCs. Note: the color scale indicates the normalized information, red represents high values and blue represents low values. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

number of spectral and image information. However, they could only describe the feature of tea samples from one aspect. The spectral data could describe the internal component information of fermentation samples. The image data could describe color and texture spatial information of tea samples. Hence, the spectral and image information should be extracted from raw hyperspectral image respectively. For the extraction of sample spectra, the average spectra of all pixels in the tea sample region were extracted and these data were processed by Z-score method. For the image information, three single-band grayscale images including 449.33 nm (B), 553.07 nm (G) and 658.37 nm (R) were selected to synthesize a color image because this color image was considerable similar to the fermentation samples (An, Huang et al., 2022). Before feature extraction, all the tea sample region of each synthesized fermentation images was considered as one region of interest (ROI), which was marked using the MATLAB GUI module (copyright no.: 2013SR122183). Subsequently, 12 color features including red component mean value(R), green component mean value(G), blue component mean value(B), hue mean value(H), saturation mean value (S), visible light mean value(V), brightness component mean value(L*), a component mean value(a*), b component mean value(b*), ultragreen transform (2G-R-B), ratio of red component mean value to green component mean value (R/G), color angle (hab*) and 6 texture features, such as average gray value (m), standard deviation (δ), smoothness (r), third moment (μ), consistency (U) and entropy (e), were extracted based on selected ROI region and this module. The detailed extraction process of these features and the software have been described in previous studies (Dong, Liang et al., 2018; Zhu et al., 2017). Finally, these extracted features were processed by normalization method. These extracted features were used to establish single information prediction models and fused information prediction models (shown in S-Fig. 2).

#### 2.5.2. Data fusion and effective information selection

In this study, three different information sources including spectra, image and electrical properties, described the characteristics of fermentation samples from different aspects and they expressed relatively independent sample information. Hence, data fusion strategy should be developed to obtain more comprehensively sample information from different aspects. The fusion information contained the internal component, color and spatial texture information of accumulated samples, indicating that data fusion strategy could obtain relatively comprehensive sequential fermentation samples information. In this research, the preprocessed data from three different information source were concatenated into a new matrix (also called low-level fusion), thereby representing the comprehensive sample information. Nevertheless, the new matrix contained a large number redundant information, which could be explained by other typical characteristics. In addition, the irrelevant information would not only reduce the accuracy of the model, but also affect the running speed of the program. Hence, some effective information selection method should be developed. To improve the accuracy and running speed of the data fusion model, the informative features of the raw data were extracted from each single data source to form a new matrix. The Pearson correlation analysis was used to select the effective color and texture variables for the image information, whereas the competitive adaptive reweighted sampling (CARS) (Li & Chen, 2017), bootstrapping soft shrinkage (BOSS) (Deng et al., 2016) and model adaptive space shrinkage (MASS) (Wen et al., 2016) were applied to select effective spectral bands and electrical parameters. The data fusion and effective features selection were displayed in Fig. 1 (d).

#### 2.5.3. Establishment and evaluation of regression models

In this study, the classical linear partial least squares regression (PLSR) algorithm was applied to establish the major chemical components prediction models based on single data and fusion data, respectively. The classical nonlinear support vector regression (SVR) algorithm was used to establish 5-fold cross validation model for major chemical components using fusion information. Subsequently, the stacking combination strategy based on fusion data was proposed.

Stacking ensemble is a typical strategy of heterogeneous combination in ensemble learning, which combined the individual generalizers in decision level to improve the generalization and robustness of the model. In this study, a two-layer stacking combination model framework was developed to evaluate the fermentation quality of black tea. The proposed stacking combination strategy was displayed in Fig. 1 (e). For the first layer, the classical PLSR and nonlinear SVR model were applied to obtain the 5 cross-validation results of fusion information. For the second layer, the obtained cross-validation results were input into the random forest (RF) model to evaluate the fermentation quality of the black tea.

In order to evaluate the model performance, some noteworthy parameters, such as the correlation coefficient of the calibration set ($R_c$), the correlation coefficient of the cross-validation set ($R_{cv}$), the correlation coefficient of the prediction set ($R_p$), the root mean square error of calibration set (RMSEC), cross-validation set (RMSECV) and the prediction set (RMSEP), the radio of cross-validation to deviation (RVD) and the radio of prediction to deviation (RPD), were applied to evaluate the performance of these established models. Generally, the higher value of $R_c$, $R_{cv}$, $R_p$, RVD and RPD, the better performance of our proposed model. Remarkably, in our study, all samples were carried out 5 cross-validation, thereby the $R_{cv}$ and RVD value could be compared with the $R_p$ and RPD value, respectively.

### 2.6. Software

The statistical graphs were drawn using Origin 2021b (OriginLab Corp. Massachusetts, USA). All the data analyses were carried out by Matlab R2017b software (The Math Work, Inc., Natick, MA, USA).

## 3. Results and discussion

### 3.1. Trend of major chemical components over fermentation time

In this research, the total catechin, soluble sugar and caffeine were considered as the major chemical components during black tea fermentation because they were closely related to the sensory quality of black tea and the trend of these major components over fermentation time was demonstrated in Fig. 2 (a). Overall, the content of total catechin decreased over time. During black tea fermentation, the catechin showed strong enzymatic oxidation reactions to form some water-soluble tea pigments, such as theaflavins (TF), thearubigins (TR) and theabrownin (TB), and some volatile aromatic compounds, resulting in changes in color and aroma of fermentation samples (Li et al., 2021). Hence, catechins play an extremely important role in the special color and aroma formation of black tea. The soluble sugar displayed a trend of decreasing first, subsequently increasing and then decreasing over fermentation time. The soluble sugar mainly contains monosaccharides and polysaccharides. The monosaccharides were applied for the respiration of fermentation leaves, whereas the polysaccharides were hydrolyzed into monosaccharide. When the content of polysaccharides hydrolysis exceeded the consumption of monosaccharide respiration, the soluble sugar showed an increasing trend, leading to a significant increase in the sweetness of tea infusion. Generally, this phenomenon occurs in the stage of moderate fermentation. Hence, in this study, the soluble sugar showed an increasing trend at 3 h and 4 h for black tea fermentation. Subsequently, the monosaccharides were consumed by excessive respiration, representing a downward trend when the excessive fermentation reached. In contrast to catechin and soluble sugar, the caffeine did not change obviously corresponded to its stable chemical properties (Yang et al., 2021).

### 3.2. Feature analysis of single digital information

In order to evaluate the fermentation quality, three digital information including spectra, image and electrical properties were applied to evaluate the major chemical components. They described the tea samples from different aspects, and they need to be further analyzed respectively.

#### 3.2.1. Response spectra analysis

The Fig. 1 (c-1 and 2) displayed the change of sample spectra in the range of 400–1000 nm with the fermentation time and degree. Obviously, the average spectra of fermentation samples displayed the same trend in the range of 400–1000 nm. Nevertheless, the reflectance of the sample spectra showed obvious difference, especially in the range of 780–1000 nm, and they might be attributable to the stretching and vibration of the third and fourth overtone of group C—H (Li, Jin, Sun et al., 2019). Although the difference of the spectral reflectance is not obvious in the visible range, some small absorption peaks still show dynamic changes with the fermentation time, which might be related to the change of sample color. Hence, more effective spectral information should be further selected and analyzed.

#### 3.2.2. Color and texture features analysis

All the obtained image data were applied to extract 18 color and texture features, which displayed some changes from the perspective of human vision, especially for the color features (shown in Fig. 1 (c-3)). Obviously, the overall color of tea samples changed from dark green to yellowish-brown first and then to dark brown as fermentation progressed, the phenomenon was related to the damage of the palisade tissue and the formation of tea pigment. To further explore color and texture features, the Pearson correlation analysis was performed between each feature, which was shown in Fig. 1 (c-4). Most variables exhibited a high correlation with other variables. Hence, these color and texture features contained some redundant information, which should be further eliminated.

#### 3.2.3. Electrical parameters analysis

As can be seen in Fig. 2 (b)-(h), the changing trend of 7 electrical parameters after preprocessed over fermentation time and test frequency were displayed. In Fig. 2 (b), the Lp represented relatively special characteristics over time and frequency. The Lp gradually decreased over fermentation time and gradually increased when the test frequency was lower than 0.4 kHz. The Lp remained stable when the test frequency higher than 0.4 kHz. According to Fig. 2 (c) and (f), the Rp and Z decreased with the increase frequency but remained relatively stable over fermentation time. As can be seen in Fig. 2 (d), (e) and (g), the regular patterns of the D, Q and θ of tea samples were similar with the increase of fermentation time. However, there were still some subtle differences in the regular patterns of these parameters with the increase of test frequency. When the test frequency was higher than 15 kHz, the Q did not change at the same fermentation time. However, the D displayed a tardy downward trend first and then tended to be constant with the increase of test frequency. In contract to the parameter D, the regular patterns of the parameter θ showed a tardy increasing trend. According to Fig. 2 (h), when the test frequency was lower than 15 kHz, the X gradually increased over fermentation time and increased first and then decreased with the increase of test frequency at the initial fermentation stage. Subsequently, the X displayed a downward trend with the increase of test frequency. When the test frequency was higher than 15 kHz, the X did not display obvious change over time but gradually increased with the increase of test frequency. During black tea fermentation, the change in cell architecture and constituents would affect the ability of molecules to capture charge. Therefore, the above electrical parameters displayed a significant changing trend.

#### 3.2.4. Hierarchical clustering analysis (HCA) of single digital information

To further explore the response ability of data information over fermentation time, these data information were subject to principal component analysis (PCA) and then the first 15 PCs were applied for HCA based on the Euclidean distance. Because the contribution rates of the first 15 PCs for spectra, image and electrical parameters were 99.99%, 99.96% and 100% respectively, they represent almost all the sample information. According to Fig. 2 (a), these components showed a certain trend with fermentation time. Therefore, the samples were divided into eight categories based on the fermentation process. As can be seen in Fig. 2 (i)-(k), the HCA for spectra, image and electrical parameters were displayed respectively. They presented poor clustering results, i.e., some adjacent categories were incorrectly divided. Importantly, according to Fig. 2 (k), the first 15 PCs of electrical parameters could accurately identify the black tea samples of initial fermentation, indicating that electrical properties technology had good discrimination ability for initial fermentation samples. However, the HCA is unsupervised analysis method, which could not predict the composition content of unknown samples. To achieve quantitative prediction of some major components in black tea fermentation, some classical regression algorithms should be applied.

### 3.3. Effective feature selection for each single digital information

The raw data contain a large number of redundant information and the processing of multidimensional fusion data is time-consuming. Hence, the prediction model established by multidimensional information is difficult to meet the requirements of black tea actual production. In order to meet the requirements of black tea actual production, the effective features of raw data should be extracted from each single digital information. In this study, the CARS, BOSS and MASS strategy were applied to select effective features for spectral and electrical parameters, and the Pearson correlation analysis was used to eliminate irrelevant color and texture features.

#### 3.3.1. Feature selection for spectra

In CARS, the number of Monte Carlo Sampling runs, the extraction of maximal principle and the group number for cross validation were 100, 18 and 5, respectively. Subsequently, three groups of band sets including 101 bands for total catechins, 107 for soluble sugar and 76 for caffeine were obtained. In BOSS, the number of bootstrap sampling and the maximal number of latent variables were 1000 and 18, respectively. Some effective band sets including 73 bands for total catechins, 97 bands for soluble sugar and 92 bands for caffeine were selected. In MASS, the group number for cross validation and the number of binray matrix sampling were 5 and 2000, respectively. Some effective features, i.e., 220 bands for total catechins, 59 bands for soluble sugar and 99 bands for caffeine were obtained. These selected bands for total catechins, soluble sugar and caffeine were represented in Fig. 3 (a)-(c). Based on these feature selection strategies, total catechins obtained the largest number of effective bands, which might be related to the considerable changes in the concentration of catechin. In addition, most effective bands were located in the range of 780–1000 nm, which belonged to near infrared region and was attributable to the stretching and vibration of the third and fourth overtone of group C—H (Li et al., 2019).

#### 3.3.2. Feature selection for color and texture information

In this study, 18 color and texture features were collected to express the external information of tea samples. There were three different research objectives, which displayed different regular patterns. Therefore, the Pearson correlation analysis was applied to select effective color and texture features for three different research objectives. According to Fig. 3 (d)-(f), some effective features were selected. Although these features did not display high correlation coefficients, they were statistically significantly related to these major compositions. For total catechins, 13 effective features including the G, B, H, S, L*, a*, b*, 2G-R-

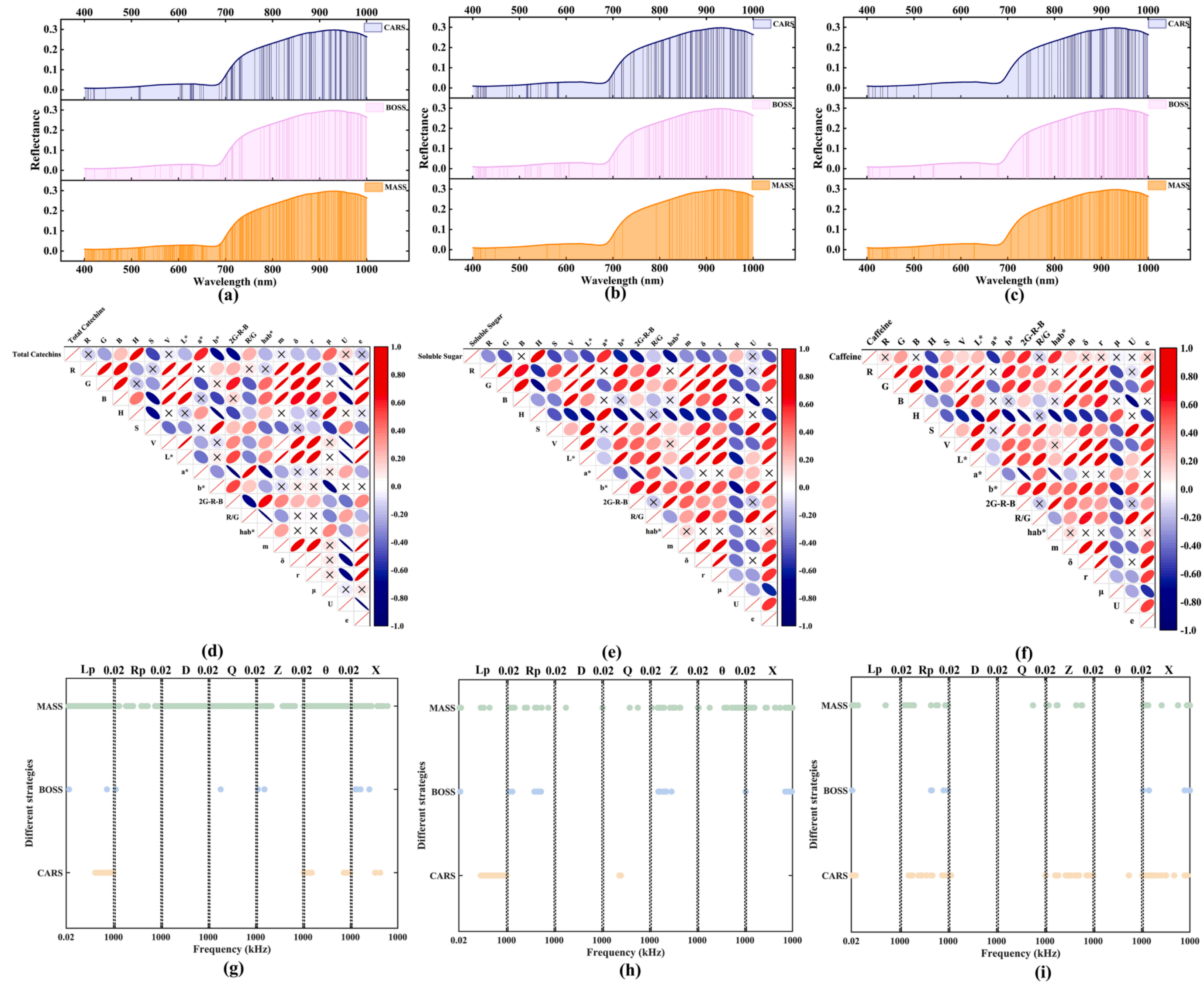


**Fig. 3.** The spectral effective features selection for (a) total catechins, (b) soluble sugar and (c) caffeine. The effective color and texture features selection for (d) total catechins, (e) soluble sugar and (f) caffeine. The effective electrical parameters selection for (g) total catechins, (h) soluble sugar and (i) caffeine.

B, R/G, hab*, δ, r and μ were retained and the correlation coefficients of these features were −0.26, 0.22, 0.79, −0.52, −0.22, 0.59, −0.69, −0.69, 0.25, −0.20, −0.19, −0.18 and 0.46, respectively. For soluble sugar, 16 color and texture features, i.e., R, G, H, S, V, L*, a*, b*, 2G-R-B, R/G, hab*, m, δ, r, μ and e, were selected and the correlation coefficients were −0.32, −0.45, 0.74, −0.50, −0.31, −0.49, 0.57, −0.58, −0.66, −0.16, −0.65, −0.31, −0.43, −0.35, 0.21 and −0.45, respectively. In contrast to total catechins and soluble sugar, only 10 variables, such as G, H, S, V, L*, a*, b*, 2G-R-B, hab* and m, exhibited a significant correlation with the change of caffeine. The correlation coefficients were 0.30, −0.54, 0.25, 0.16, 0.23, −0.60, 0.35, 0.60, 0.58 and 0.16, respectively. Obviously, more color features were selected than texture features because of the transformation of catechins and the formation of tea pigment.

### 3.3.3. Feature selection for electrical parameters

For these feature selection algorithms, the key parameters are constant. The selected effective electrical parameters for total catechins, soluble sugar and caffeine were displayed in Fig. 3 (g)-(i), respectively. For total catechins, the selected electrical parameters using CARS, BOSS and MASS were 48, 11 and 334, respectively. Importantly, the Lp at high frequency were selected by three feature selection algorithms simultaneously, indicating that the Lp was an essential feature to describe the change of total catechins. For the soluble sugar, the CARS, BOSS and MASS strategies selected 37, 25 and 70 electrical parameters, respectively. However, these three algorithms did not choose common electrical parameters, indicating that each electrical parameter has a unique role in expressing the change of soluble sugar. For the caffeine, some effective electrical parameters including 63 using CARS, 10 using BOSS and 43 using MASS were obtained. Importantly, the X, Rp at high frequency and Lp at low frequency were selected by three feature selection algorithms simultaneously, showing that these parameters play an important role in expressing the change of caffeine.

## 3.4. Quantitative prediction model with single digital information

The PLSR model was applied to predict these major components, i.e., total catechins, soluble sugar and caffeine, based on single digital information with full features during black tea fermentation. Before modeling, all the fermentation samples were divided into calibration set (107 samples) and prediction set (53 samples) based on the Kennard-Stone method with the radio of 2:1. In Table 1, the established PLSR

**Table 1**
The PLSR models for major chemical composition of fermentation leaves based on individual data.

| Chemical composition | Data | LVs | Calibration set | | Prediction set | |
|---|---|---|---|---|---|---|
| | | | $R_c$ | RMSEC | $R_p$ | RMSEP |
| Total catechins | Spectra | 5 | 0.9045 | 0.5823 | 0.9164 | 0.5644 |
| | Image | 8 | 0.8895 | 0.6851 | 0.8770 | 0.6971 |
| | Electric | 5 | 0.8803 | 0.7695 | 0.8878 | 0.7757 |
| Soluble sugar | Spectra | 5 | 0.9068 | 0.3643 | 0.9131 | 0.3503 |
| | Image | 10 | 0.8606 | 0.4400 | 0.8679 | 0.4498 |
| | Electric | 4 | 0.8782 | 0.4133 | 0.8896 | 0.3929 |
| Caffeine | Spectra | 5 | 0.7299 | 0.1015 | 0.7717 | 0.1136 |
| | Image | 11 | 0.7425 | 0.0995 | 0.7060 | 0.1177 |
| | Electric | 5 | 0.7191 | 0.1032 | 0.8365 | 0.0949 |

model for total catechins and soluble sugar using single information represented similar prediction performance, which might be related to the large change gradient of the total catechins and soluble sugar. Compared with these models, the established models for caffeine represented poor prediction performance. For the total catechins and soluble sugar, the established model using spectral data displayed the best performance and the prediction model for caffeine using electrical parameters showed the best performance, indicating that different detection technologies had different representational ability for these major chemical components. In addition, the performance of established prediction models had high potential for improvement. Hence, different detection technologies should be combined to obtain more comprehensive information for fermentation samples to improve the performance of the prediction model for major chemical components.

### 3.5. Quantitative prediction model with fusion information

To obtain more comprehensive sample information and establish more accurate prediction model, the data fusion strategy was developed. In this study, all obtained information using different detection technology were concatenated into a new matrix, which was subjected to normalization for processing. Subsequently, the quantitative prediction models for major chemical components using fusion information were established. In addition, in order to meet the requirements of black tea processing in actual production, some effective features were selected from each single digital information to form a new matrix for the establishment of major chemical components based on our proposed stacking combination strategy.

#### 3.5.1. Establishment of 5-fold cross validation model with full features

In order to more accurately evaluate the performance of the prediction model, the classical linear PLSR model and nonlinear SVR model were carried out 5-fold cross validation and the results were displayed in S-Table 1. Because the results of cross validation for each fold were easily affected by uneven sample division, the results of cross validation for all samples were compared and analyzed. For the total catechins and caffeine, the PLSR and SVR model exhibit similar results. However, for the soluble sugar, the SVR model showed better prediction performance, which might be caused by complicated changes of soluble sugar, such as the respiration of monosaccharides and the degradation of polysaccharides. Although the established prediction model using fusion information showed better performance than the established model using single digital information, their performance still had the potential for further improvement, especially for the caffeine. Hence, the novel modeling strategy that can improve the robustness and accuracy of the prediction model need to be performed.

#### 3.5.2. Establishment of stacking combination model with full features

To improve the robustness and accuracy of the prediction model, the stacking combination strategy was developed in this research. The stacking combination strategy establish the prediction model based on the decision results of each single model. The framework and performance of our proposed stacking combination strategy were displayed in Fig. 1 (e), (f) and Table 2, respectively. Remarkably, for the second layer, the results from RF model were calculated 50 times and averaged, and then they were recorded in Table 2. The RPD values for the total catechins, soluble sugar and caffeine were 10.0215, 8.4981 and 2.9258, respectively. Compared with the performance of 5-fold cross validation strategy using PLSR and SVR models, our proposed two-layers stacking combination strategy significantly improved the robustness and accuracy of the prediction model. Although our proposed stacking combination strategy achieved a satisfactory result, the calculation of the prediction model is extremely time-consuming. Hence, the information from different data source should be carried out feature selection strategy to eliminate redundant information and reduce the calculation time of model, and then these selected features were concatenated into a new matrix for the establishment of prediction model.

#### 3.5.3. Establishment of stacking combination model with effective features

To simplify the prediction model, the effective feature selection strategies including CARS, BOSS, MASS and correlation analysis were performed. The selected variables for each data source and the performance of established stacking combination models were displayed in Fig. 3 and Table 2, respectively. According to Table 2, the performance of most stacking combination models using effective features was better than that of the stacking combination models based on only fusion information, except for the total catechin model using MASS algorithm, indicating that the MASS algorithm removed some effective variables corresponding to total catechins, which affected the performance of the prediction model. The most accurate results were obtained for the prediction of total catechins based on the stacking combination model and CARS effective variables selection algorithm, with $R_p$, RMSEP and RPD being 0.9978, 0.1770 and 14.8129, respectively. For the soluble sugar, the same strategy as total catechins obtained the most accurate results, with $R_p$, RMSEP and RPD being 0.9973, 0.0631 and 13.4377, respectively. In contract to established model for total catechins and soluble sugar, the most accurate results for caffeine were obtained using the stacking combination model and MASS effective variables selection algorithm. The $R_p$, RMSEP and RPD were 0.9550, 0.0496 and 3.1824, respectively. These established models using stacking combination strategy with effective variables could accurately predict the major chemical components during black tea fermentation. This phenomenon might be caused by the following reasons. First of all, the collected information from different data source could effectively represent the change of fermentation samples. Secondly, the applied feature selection methods retained most effective variables, which was closely related to these major chemical components. Finally, our established stacking combination model had good robustness and accuracy.

### 3.6. Discussion

Although the accuracy is an important factor to evaluate the performance of the prediction model, the calculation time of the model is still crucial for the real-time detection of black tea quality. Generally speaking, the more the number of variables, the longer the calculation time of the prediction model. Hence, the performance of the prediction model should be comprehensively evaluated. The performance comparison of some significant established models was visualized in Fig. 4 (a)-(c), which represented the model performance for total catechins, soluble sugar and caffeine, respectively. Each dot corresponds to one model, the $R_p$ and the RPD value on the prediction set provide the coordinates of the dot center. The radius represents the RMSEP value of the prediction set and the number in Fig. 4 denotes the number of variables, which participate in the establishment of the prediction model. If the dot is far from the origin, the radius and the number are smaller, the corresponding model display better performance. For the total catechins, as can be seen in Fig. 4 (a), the stacking fusion-CARS and the stacking

**Table 2**
The stacking model for major chemical composition of fermentation leaves based on data fusion and information selection strategy.

| Chemical composition | Methods | Parameter | Calibration set | | Prediction set | | |
|---|---|---|---|---|---|---|---|
| | | | $R_c$ | RMSEC | $R_p$ | RMSEP | RPD |
| Total catechins | fusion | tree = 110, mtry = 2 | 0.9985 | 0.1443 | 0.9956 | 0.2557 | 10.0215 |
| | Fusion-CARS | tree = 200, mtry = 13 | 0.9989 | 0.1231 | 0.9978 | 0.1770 | 14.8129 |
| | Fusion-BOSS | tree = 110, mtry = 18 | 0.9983 | 0.1544 | 0.9978 | 0.1798 | 14.5465 |
| | Fusion-MASS | tree = 30, mtry = 1 | 0.9981 | 0.1623 | 0.9939 | 0.2876 | 9.2827 |
| Soluble sugar | fusion | tree = 100, mtry = 14 | 0.9980 | 0.0549 | 0.9932 | 0.1004 | 8.4981 |
| | Fusion-CARS | tree = 60, mtry = 14 | 0.9989 | 0.0409 | 0.9973 | 0.0631 | 13.4377 |
| | Fusion-BOSS | tree = 190, mtry = 20 | 0.9980 | 0.0547 | 0.9947 | 0.0892 | 9.8552 |
| | Fusion-MASS | tree = 300, mtry = 19 | 0.9977 | 0.0585 | 0.9936 | 0.0977 | 8.9711 |
| Caffeine | fusion | tree = 20, mtry = 9 | 0.9807 | 0.0297 | 0.9488 | 0.0525 | 2.9258 |
| | Fusion-CARS | tree = 370, mtry = 14 | 0.9864 | 0.0247 | 0.9511 | 0.0517 | 2.9947 |
| | Fusion-BOSS | tree = 420, mtry = 2 | 0.9903 | 0.0209 | 0.9560 | 0.0501 | 3.1424 |
| | Fusion-MASS | tree = 120, mtry = 12 | 0.9876 | 0.0237 | 0.9550 | 0.0496 | 3.1824 |

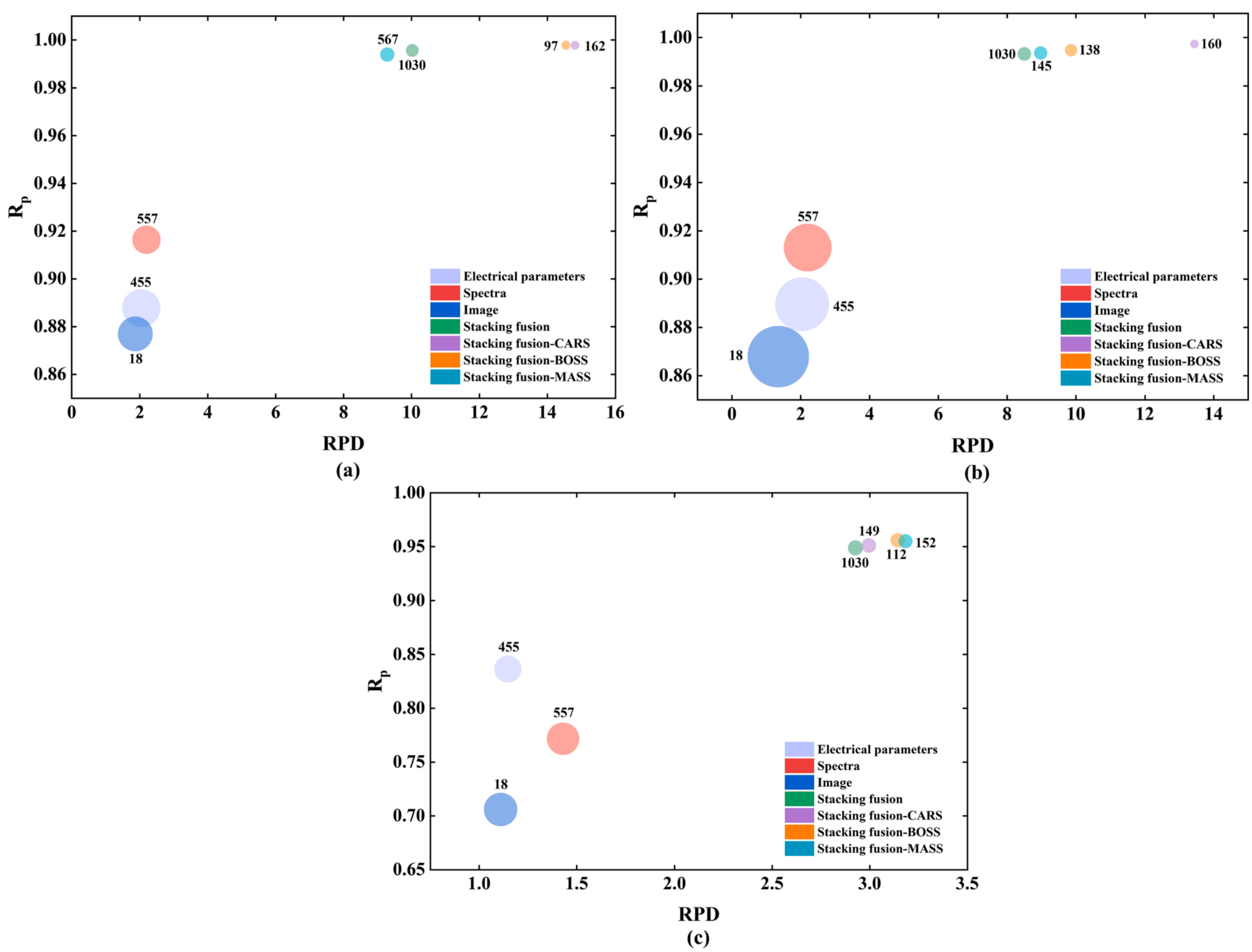


**Fig. 4.** The comparison of the stacking combination model and other models: (a) total catechins, (b) soluble sugar and (c) caffeine.

fusion-BOSS exhibited similar prediction accuracy. If some established models display similar prediction accuracy, we will choose the model with fewer variables because fewer variables can reduce the calculation time of the stacking fusion model and help achieve real-time detection of major components for black tea in actual production. In Fig. 4 (c), the same reasons are true for the selection of best stacking fusion model for caffeine. In Fig. 4 (b), the stacking fusion-CARS model demonstrates significant advantages compared with other established models. After comprehensive consideration of the factors affecting the model, such as the accuracy and calculation time of the prediction model, the following conclusions were obtained. For the total catechins and caffeine, the stacking fusion-BOSS strategy were considered to be the best model. For the soluble sugar, the best performance was displayed by stacking fusion-CARS strategy. From the distribution of these dots representing the model performance in Fig. 4, it can be seen that the stacking combination model using fusion data displayed better predictive performance than the established model using single data. This phenomenon indicated that our proposed multielement information including spectra, image and electrical parameters were complementary. In addition, the selection of effective features and the fusion of decision results also helped the model make better decisions on given targets.

Different technologies applied to the evaluation of sample quality during black tea processing were compared. According to S-Table 2, some single technologies including VIS-NIR spectra (Yang et al., 2021), CVS (Dong, Li et al., 2018) and electrical properties (Dong et al., 2021) have been applied to quantitatively evaluate the fermentation quality of black tea. However, these single technologies exist certain limitations. These single technologies could only describe the characteristics of tea samples from one aspect. The VIS-NIR spectra and CVS technology could detect components changes based on the response of some characteristic groups and the change of color and texture features. Nevertheless, the electrical properties represented the sample information according to the ability of molecules to capture charges because the change in cell architecture and constituents would improve the reactance and reduce the capacity of tolerance. Remarkably, black tea fermentation involved some complicated oxidation and reduction reaction, resulting in a considerable change of the internal and external features for fermentation samples. Hence, more comprehensive sample information should be collected from different aspects to evaluate the black tea fermentation quality. Wang et al., successfully evaluated the tea quality using the fusion information of NIRS and CVS technology during black processing (Wang, Li, Liu et al., 2021). Based on the same fusion information, Jin et al., predicted the pigments content during black tea fermentation, with RPD for TF, TFDG, TF-3-G and TF-3′-G being 2.01, 0.99, 1.36, and 1.18, respectively (Jin et al., 2021). However, some components, such as TFDG, TF-3-G and TF-3′-G, would not be accurately predicted, this may be because incomplete information was obtained by NIRS and CVS technology, whose penetration ability were limited. Generally, a small number of samples would be unrepresentative and moderate samples would display a certain thickness. In order to obtain overall information of the collected samples, more detection technology should be applied and fused to evaluate the fermentation quality of black tea. Remarkably, the electrical properties technology regarded the selected sample as a complex of dielectrics, electrolytes and conductors, indicating that it could obtain the electrical parameters of whole selected samples. Therefore, in this study, the spectra, image and electrical parameters were applied to describe the sample feature to obtain the comprehensive sample information. To the best of our knowledge, this is the first study to use the fusion data of three different detection information, such as spectra, image and electrical properties, for predicting major components corresponding to the taste of finished tea during black tea fermentation. In addition, this is also the first study to apply the stacking combination strategy for evaluating the fermentation quality of black tea in decision level.

Although our research achieves a satisfactory result, some defects still exist and need to be improved, including the limitations of seasons and sample varieties and the application of multielement information collection system. Hence, our further studies include that (1) more fermentation experiments should be carried out using different tea varieties in different seasons, (2) the low cost multielement information, including hyperspectral image and electrical properties, collection instrument should be developed for the intelligent evaluation of black tea quality.

## 4. Conclusion

In this study, using multielement information fusion method and stacking combination strategy for quantitative evaluating the fermentation quality of black tea was demonstrated to be feasible. The quantitative prediction was made for three major chemical components including total catechins, soluble sugar and caffeine. Compared with the established models using single information, the multielement information fusion models displayed better prediction performance for total catechins, soluble sugar and caffeine. In addition, the established stacking combination models using effective features achieved more satisfactory prediction results at the decision level, with RPD for total catechins, soluble sugar and caffeine being 14.5465, 13.4377 and 3.1424, respectively. Our study provides a novel method for quantitative detection of major components corresponding to the taste of finished tea in black tea fermentation.

## CRediT authorship contribution statement

**Ting An:** Conceptualization, Methodology, Investigation, Writing – original draft. **Zheli Wang:** Methodology, Investigation, Visualization. **Guanglin Li:** Investigation, Methodology, Formal analysis. **Shuxiang Fan:** Validation. **Wenqian Huang:** Data curation, Formal analysis, Visualization. **Dandan Duan:** Project administration. **Chunjiang Zhao:** Resources, Project administration, Funding acquisition. **Xi Tian:** Methodology, Formal analysis, Funding acquisition. **Chunwang Dong:** Conceptualization, Project administration, Funding acquisition.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

The data that has been used is confidential.

## Acknowledgments

This study was supported by National Natural Science Foundation of China (31972466). Research and development of precision detection technology and equipment for fruit quality in Beijing, China (CZZJ202203). The Research start-up funds-TRI-SAAS of China (CXGC2023F18, CXGC2023G33). Remote sensing evaluation and application of tea quality and tea garden disaster in mountainous areas in China (2021YFD1601103). Project funded by China Postdoctoral Science Foundation (323241).

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2023.100718.

## References

An, T., Huang, W., Tian, X., Fan, S., Duan, D., Dong, C., … Li, G. (2022). Hyperspectral imaging technology coupled with human sensory information to evaluate the fermentation degree of black tea. *Sensors and Actuators B: Chemical, 366*. https://doi.org/10.1016/j.snb.2022.131994

An, T., Li, Y., Tian, X., Fan, S., Duan, D., Zhao, C., … Dong, C. (2022). Evaluation of aroma quality using multidimensional olfactory information during black tea fermentation. *Sensors and Actuators B-Chemical, 371*. https://doi.org/10.1016/j.snb.2022.132518

An, T., Yu, S., Huang, W., Li, G., Tian, X., Fan, S., … Zhao, C. (2022). Robustness and accuracy evaluation of moisture prediction model for black tea withering process using hyperspectral imaging. *Spectrochim Acta A Mol Biomol Spectrosc, 269*, Article 120791. https://doi.org/10.1016/j.saa.2021.120791

Chen, Q., Chen, M., Liu, Y., Wu, J., Wang, X., Ouyang, Q., & Chen, X. (2018). Application of FT-NIR spectroscopy for simultaneous estimation of taste quality and taste-related compounds content of black tea. *Journal of Food Science and Technology-Mysore, 55* (10), 4363–4368. https://doi.org/10.1007/s13197-018-3353-1

Deng, B., Yun, Y., Cao, D., Yin, Y., Wang, W., Lu, H., … Liang, Y. (2016). A bootstrapping soft shrinkage approach for variable selection in chemical modeling. *Anal Chim Acta, 908*, 63–74. https://doi.org/10.1016/j.aca.2016.01.001

Dong, C., Li, J., Wang, J., Liang, G., Jiang, Y., Yuan, H., … Meng, H. (2018). Rapid determination by near infrared spectroscopy of theaflavins-to-thearubigins ratio during Congou black tea fermentation process. *Spectrochim Acta A Mol Biomol Spectrosc, 205*, 227–234. https://doi.org/10.1016/j.saa.2018.07.029

Dong, C., Liang, G., Hu, B., Yuan, H., Jiang, Y., Zhu, H., & Qi, J. (2018). Prediction of Congou Black Tea Fermentation Quality Indices from Color Features Using Non-

Linear Regression Methods. *Sci Rep, 8*(1), 10535. https://doi.org/10.1038/s41598-018-28767-2

Dong, C., Ye, Y., Yang, C., An, T., Jiang, Y., Ye, Y., … Yang, Y. (2021). Rapid detection of catechins during black tea fermentation based on electrical properties and chemometrics. *Food Bioscience, 40*. https://doi.org/10.1016/j.fbio.2020.100855

Fan, S., Liang, X., Huang, W., Zhang, V., Pang, Q., He, X., … Zhang, C. (2022). Real-time defects detection for apple sorting using NIR cameras with pruning-based YOLOV4 network. *Computers and Electronics in Agriculture, 193*. https://doi.org/10.1016/j.compag.2022.106715

Jin, G., Wang, Y., Li, L., Shen, S., Deng, W., Zhang, Z., & Ning, J. (2020). Intelligent evaluation of black tea fermentation degree by FT-NIR and computer vision based on data fusion strategy. *Lwt, 125*. https://doi.org/10.1016/j.lwt.2020.109216

Jin, G., Wang, Y., Li, M., Li, T., Huang, W., Li, L., … Ning, J. (2021). Rapid and real-time detection of black tea fermentation quality by using an inexpensive data fusion system. *Food Chem, 358*, Article 129815. https://doi.org/10.1016/j.foodchem.2021.129815

Li, J., & Chen, L. (2017). Comparative analysis of models for robust and accurate evaluation of soluble solids content in 'Pinggu' peaches by hyperspectral imaging. *Computers and Electronics in Agriculture, 142*, 524–535. https://doi.org/10.1016/j.compag.2017.11.019

Li, L., Li, M., Liu, Y., Cui, Q., Bi, K., Jin, S., … Zhang, Z. (2021). High-sensitivity hyperspectral coupled self-assembled nanoporphyrin sensor for monitoring black tea fermentation. *Sensors and Actuators B: Chemical, 346*. https://doi.org/10.1016/j.snb.2021.130541

Li, L., Wang, Y., Jin, S., Li, M., Chen, Q., Ning, J., & Zhang, Z. (2021). Evaluation of black tea by using smartphone imaging coupled with micro-near-infrared spectrometer. *Spectrochim Acta A Mol Biomol Spectrosc, 246*, Article 118991. https://doi.org/10.1016/j.saa.2020.118991

Li, X., Jin, J., Sun, C., Ye, D., & Liu, Y. (2019). Simultaneous determination of six main types of lipid-soluble pigments in green tea by visible and near-infrared spectroscopy. *Food Chemistry, 270*, 236–242. https://doi.org/10.1016/j.foodchem.2018.07.039

Qu, F., Zeng, W., Tong, X., Feng, W., Chen, Y., & Ni, D. (2020). The new insight into the influence of fermentation temperature on quality and bioactivities of black tea. *Lwt-Food Science and Technology, 117*. https://doi.org/10.1016/j.lwt.2019.108646

Zarezadeh, M. R., & Aboonajmi, M. (2023). The Effect of Data Fusion on Improving the Accuracy of Olive Oil Quality Measurement. *Food Chemistry, X*. https://doi.org/10.1016/j.fochx.2023.100622

Wang, Y., Li, L., Liu, Y., Cui, Q., Ning, J., & Zhang, Z. (2021). Enhanced quality monitoring during black tea processing by the fusion of NIRS and computer vision. *Journal of Food Engineering, 304*. https://doi.org/10.1016/j.jfoodeng.2021.110599

Wen, M., Deng, B., Cao, D., Yun, Y., Yang, R., Lu, H., & Liang, Y. (2016). The model adaptive space shrinkage (MASS) approach: A new method for simultaneous variable selection and outlier detection based on model population analysis. *Analyst, 141*(19), 5586–5597. https://doi.org/10.1039/c6an00764c

Yang, C., Zhao, Y., An, T., Liu, Z., Jiang, Y., Li, Y., & Dong, C. (2021). Quantitative prediction and visualization of key physical and chemical components in black tea fermentation using hyperspectral imaging. *Lwt, 141*. https://doi.org/10.1016/j.lwt.2021.110975

Yu, D., Guo, S., Zhang, X., Yan, H., Zhang, Z. Y., Chen, X., … Duan, J. (2022). Rapid detection of adulteration in powder of ginger (Zingiber officinale Roscoe) by FT-NIR spectroscopy combined with chemometrics. *Food Chem X, 15*, Article 100450. https://doi.org/10.1016/j.fochx.2022.100450

Zhu, H., Liu, F., Ye, Y., Chen, L., Liu, J., Gui, A., … Dong, C. (2019). Application of machine learning algorithms in quality assurance of fermentation process of black tea– based on electrical properties. *Journal of Food Engineering, 263*, 165–172. https://doi.org/10.1016/j.jfoodeng.2019.06.009

Zhu, H., Ye, Y., He, H., & Dong, C. (2017). Evaluation of green tea sensory quality via process characteristics and image information. *Food and Bioproducts Processing, 102*, 116–122. https://doi.org/10.1016/j.fbp.2016.12.004