

DETECTION OF FLAXSEED OIL ADULTERATION BASED ON TWO-DIMENSIONAL CORRELATION NEAR-INFRARED SPECTRA

基于二维相关近红外光谱的亚麻籽油掺杂检测

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ABSTRACT

Flaxseed oil is rich in α -linolenic acid and other nutrients, and the adulteration happens frequently because of its high price. To detect the adulteration of flaxseed oil quickly and accurately, a method was proposed based on weighted reconstructed two-dimensional correlation near-infrared (NIR) spectra. The near-infrared spectra of 79 adulterated flaxseed oil samples were measured, and the traditional two-dimensional correlation synchronous spectra were calculated. The two-dimensional correlation synchronous spectra of all samples were decomposed into multiple components of different scales by the bi-dimensional empirical mode decomposition algorithm (BEMD). According to the root mean square error (RMSE) values of the adulteration detection sub-models established by each component, the weights of the corresponding components were calculated, and then the two-dimensional correlation spectra of all samples were reconstructed by accumulating the weighted components. A quantitative analysis model of flaxseed oil adulteration was established based on the weighted reconstructed two-dimensional correlation spectra combined with the N-way partial least square (N-PLS) algorithm. Compared with the traditional two-dimensional correlation spectroscopy, the model built by the weighted reconstructed two-dimensional correlation spectra had better performance with the calibration determination coefficient increased by 6.05%, and the prediction determination coefficient increased by 7.5%. The proposed method could effectively enhance the spectral feature information, reduce the spectral noise interference, and hence provide a new idea for the detection of edible oil adulteration.

摘要

亚麻籽油富含 α -亚麻酸等营养成分, 市场价格较高, 掺假现象频出。为快速、准确检测亚麻籽油掺杂, 本文提出一种基于加权重构二维相关近红外光谱的亚麻籽油掺杂检测方法。采集了 79 个掺杂亚麻籽油样本(以菜籽油为掺杂油, 掺杂体积比例 1%-40%) 的近红外光谱, 计算各样本的常规二维相关同步光谱; 利用二维经验模态分解算法将各样本的二维相关光谱分解为不同尺度的多个分量, 以各分量所建立的掺杂检测子模型的均方根误差值为依据, 计算相应尺度分量的加权值, 然后通过加权分量叠加重构各样本的二维相关光谱; 基于加权重构的二维相关光谱结合多维偏最小二乘方法建立了亚麻籽油掺杂的定量分析模型。结果表明: 相比于常规二维相关光谱, 加权重构二维相关光谱建立的掺杂检测模型性能更佳, 模型的校正决定系数提高了 6.05%, 预测决定系数提高了 7.5%, 该方法可有效增强光谱特征信息, 降低光谱噪声干扰, 为食用油掺假检测提供了一种新的思路。

INTRODUCTION

Flaxseed oil is a high-quality edible oil obtained from flaxseed, which contains high concentrations of healthy compounds, such as α -linolenic acid and linoleic acid (Zhou et al., 2018). Flaxseed oil has various health-related benefits such as lowering blood pressure and lipids, anti-inflammatory and anti-cancer, and improving vision (Goyal A. et al., 2014; Liao et al., 2021; Xian et al., 2018). Because of its higher price than other vegetable oils, flaxseed oil can be adulterated with cheaper vegetable oils by some illegal oil suppliers, which will not only damage the legitimate rights and interests of consumers but also disturb the market order.

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Some analytical methods, such as gas chromatography, gas chromatography-mass spectrometry, liquid chromatography-mass spectrometry, have been reported to be successful (*Sun et al., 2015; Wei et al., 2018; Chen et al., 2012; Liao et al., 2018*); however, these methods are expensive, time-consuming, and laborious, and cannot be used for on-site real-time detection. Therefore, it is of great significance to study the rapid detection method of flaxseed oil adulteration.

Two-dimensional correlation near-infrared spectroscopy provides an attractive technique for the detection of food quality and safety due to its rapidity, simplicity, and low costs (*Chen J.B. et al., 2016; Wang et al., 2018; Yang et al., 2013; Zhang et al., 2010*). Compared with traditional one-dimensional near-infrared spectroscopy, two-dimensional correlation spectroscopy has a higher spectral resolution and better spectral selectivity and can enlarge subtle spectral information of the interested substance by specific external perturbations such as changes in temperature, and concentration (*Park et al., 2016; Yang et al., 2015*). Combined with suitable multivariate calibration methods, two-dimensional correlation spectroscopy has been applied in the detection of adulterated oil (*Zhang et al., 2019; Wang et al., 2020*). However, when the spectral resolution is improved by two-dimensional correlation spectroscopy, the influence of noise also increases, and thus false information caused by noise may appear, which will decrease the signal-to-noise ratio of two-dimensional correlation spectra and bring about the negative influence on the accuracy of the model (*Zhan et al., 2004; Sun et al., 2015*). The spectral signal has multi-scale characteristics and can be decomposed into multiple components of different frequency bands (*Chen D. et al., 2016; Liu et al., 2009*). The high-frequency components contain more noise information, the background interference is mostly concentrated in the low-frequency component, and the spectral characteristic information is mainly located in the middle-frequency components. But if the noise interference and spectral characteristic information are distinguished simply by the frequency, some of the useful spectral information may be lost, which will affect the performance of the spectral analysis model.

Based on the multi-scale characteristics of spectra, this paper presents a new method to detect flaxseed oil adulteration using weighted reconstructed two-dimensional correlation near-infrared spectroscopy. Firstly, the two-dimensional correlation near-infrared spectra of adulterated flaxseed oil samples were decomposed into multiple components by the bi-dimensional empirical mode decomposition algorithm. Then each component was weighted according to the root mean square error of the detection sub-model based on the corresponding component, and the two-dimensional correlation spectrum was reconstructed by the accumulation of weighted components. Finally, the detection model for flaxseed oil adulteration was constructed based on the weighted reconstructed two-dimensional correlation spectra in combination with the N-way partial least squares algorithm. The proposed method can mine the effective spectral information as much as possible, reduce noise and background interference, and enhance the contribution of spectral characteristic information to modeling by the weighted reconstruction of two-dimensional correlation spectra.

MATERIALS AND METHODS

Sample preparation

Pure rapeseed oil (Brand: Golden Arowana) was added to pure flaxseed oil (Brand: Fuyide) according to the adulterating volume ratio of 1%-40% (adulterating gradient: 0.5%) to prepare 79 adulterated flaxseed oil samples. Before spectral collection, all adulterated flaxseed oil samples were manually stirred to ensure homogeneity. The adulterated flaxseed oil samples were divided into the calibration set with 60 samples and the prediction set with 19 samples. The statistical parameters of the calculation set and prediction set are shown in Table 1.

Table 1

Statistics of the calculation set and prediction set

Item	Min.	Max.	Mean	Standard deviation
Calculation set adulterating ratio	1%	40%	20.66%	11.63%
Prediction set adulterating ratio	2%	38%	20%	11.25%

Spectral data acquisition

The near-infrared transmission spectra of all samples were collected using a Field Spec3 spectrometer (ASD company, USA) equipped with the InGaAs detector. The wavelength interval of spectral data is 1 nm, and the wavelength range is from 350 to 2500 nm.

The measurements were undertaken in a quartz cuvette with an optical path length of 10 mm. Taking the blank cuvette as the reference, the transmission spectra of all samples were collected. Fig.1 shows the acquisition system of near-infrared spectra.

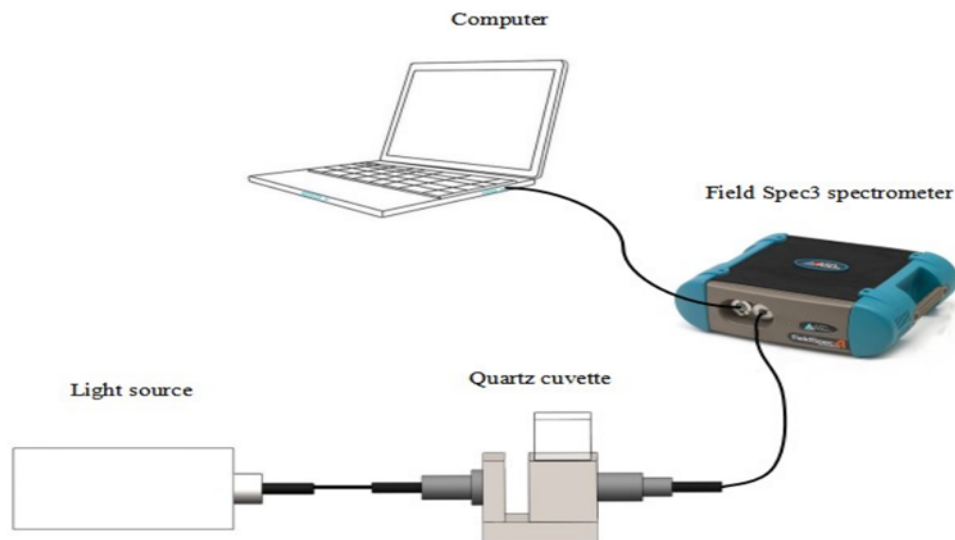


Fig. 1 - The acquisition system of near-infrared spectra

Spectral data processing

(1) Calculation of two-dimensional correlation spectra

The theory of two-dimensional correlation spectroscopy was first proposed by Noda (Noda,2004). The near-infrared transmission spectra of samples, which were measured at a specific external disturbance (different adulterating ratios of rapeseed oil), could be formed into a dynamic spectral matrix $A_{m \times n}$ with the m spectra (n wavelengths).

According to Noda's theory, two-dimensional correlation synchronous spectra of all samples could be calculated as follows:

$$\Phi(v_1, v_2) = \frac{1}{m-1} A^T A \quad (1)$$

where v_1 and v_2 represent two independent spectral wavelengths, T refers to the matrix transpose operation.

Matrix A contains 2 spectra in rows ($m=2$). The first row of A is the near-infrared transmission spectral data of the pure flaxseed oil sample, while the second row is the spectral data of the i^{th} adulterated flaxseed oil. The two-dimensional correlation near-infrared spectrum of i^{th} adulterated flaxseed oil could be obtained in terms of Eqn. (1) and by the calculation software programmed by ourselves.

(2) Weighted reconstruction of two-dimensional correlation spectra

The bi-dimensional empirical mode decomposition (BEMD) algorithm was proposed by Nune, (2005), and it can adaptively decompose the two-dimensional signals (such as image signals) into multiple components at different scales. The two-dimensional correlation spectra of all adulterated flaxseed oil samples can be decomposed by the BEMD algorithm into multiple components, which correspond to different frequency bands.

Based on the decomposed components, the detection sub-models of flaxseed oil adulteration were established respectively, in combination with the N-way partial least squares algorithm (N-PLS) and the root mean square error (RMSE) of each sub-model was calculated as follows:

$$RMSE_i = \sqrt{\frac{1}{m} \sum_{j=1}^m (y_j - y_{ij}')^2} \quad (2)$$

where:

$RMSE_i$ is the root mean square error of the sub-model built by the i^{th} component, and m is the total number of samples. y_j is the actual adulterating value of the j^{th} sample, and y_{ij}' is the predicted adulterating ratio of the j^{th} sample by the i^{th} component sub-model.

The N-PLS algorithm is a data analysis method that extends the conventional partial least squares method to high-dimensional data, which can effectively solve the modeling problem of high-dimensional data (Chu et al, 2006).

The reconstruction weight of each component is calculated based on the *RMSE* value of each sub-model as follows:

$$W_i = \frac{n \left(\frac{1}{RMSE_i} \right)}{\sum_{i=1}^n \left(\frac{1}{RMSE_i} \right)} \quad (3)$$

where: n is the total number of components of the two-dimensional correlation spectra decomposed by BEMD.

Then the two-dimensional correlation spectra of all samples can be reconstructed by accumulating the weighted components.

(3) Quantitative analysis modeling

Based on the weighted reconstructed two-dimensional correlation spectra, the quantitative analysis model of flaxseed oil adulteration was established combined with the N-PLS algorithm. Model performance was evaluated by the calibration determination coefficient (R^2_c), the prediction determination coefficient (R^2_p), the root mean square error of calibration (*RMSEC*), and the root mean square error of prediction (*RMSEP*).

These assessment parameters were calculated as follows:

$$R^2_c = 1 - \frac{\sum_{i=1}^n (\hat{y}_i - y_i)^2}{\sum_{i=1}^n (\hat{y}_i - y_c)^2} \quad (4)$$

$$R^2_p = 1 - \frac{\sum_{j=1}^n (\hat{y}_j - y_j)^2}{\sum_{j=1}^n (\hat{y}_j - y_p)^2} \quad (5)$$

$$RMSEC = \sqrt{\frac{1}{n} \sum_{i=1}^n (\hat{y}_i - y_i)^2} \quad (6)$$

$$RMSEP = \sqrt{\frac{1}{n} \sum_{j=1}^n (\hat{y}_j - y_j)^2} \quad (7)$$

where:

\hat{y}_i and y_i are the predicted and actual adulterating ratios of the i^{th} sample in the calibration set, and y_c is the averaged adulterating ratio of calibration set samples; \hat{y}_j and y_j are the predicted and actual adulterating ratios of the j^{th} sample in the prediction set, and y_p is the averaged adulterating ratio of prediction set samples; n and m are the number of samples in the calibration and prediction set, respectively.

Higher R^2_c and smaller *RMSEC* correspond to the model's high calibration accuracy, while higher R^2_p and smaller *RMSEP* correspond to high prediction stability.

All the computations above were performed in MATLAB (*Version R2015a, The Math Works Inc.*) under Windows 10.

RESULTS

Near-infrared spectra

Figure 2 showed the near-infrared spectra of pure flaxseed oil and adulterated flaxseed oil (20% adulteration ratio). Because vegetable oils are mainly composed of triglycerides and similar in chemical composition, it can be seen that the spectral waveform and absorption peak location (at 467 nm, 611 nm, 674 nm, 1208 nm, 1415 nm, 1724 nm, 1763 nm, 2144 nm, 2180 nm) of the two spectra look closely similar except for the subtle difference in absorbance. Therefore, it is difficult to carry out adulteration detection only using traditional one-dimensional spectroscopy. The spectral noise in the 2250-2500 nm band is strong and cannot provide useful information, so the spectral data in the 350-2249 nm band is intercepted for subsequent two-dimensional correlation and quantitative detection analysis.

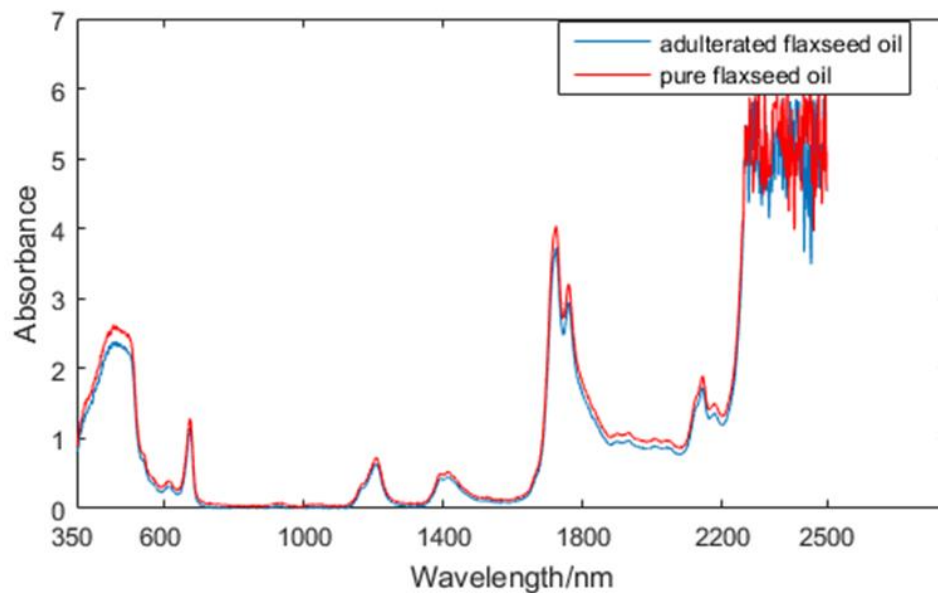


Fig. 2 - Near-infrared spectra of pure and adulterated flaxseed oil

Two-dimensional correlation spectra

Based on the collected near-infrared spectra, the two-dimensional correlation synchronous spectra of all adulterated flaxseed oil samples were calculated in terms of Eq. (1). Figure 3 showed the calculated two-dimensional correlation synchronous spectra of the flaxseed oil sample with the 20% adulterating ratio. To reduce the operation amount, the wavelength interval of the two-dimensional correlation is taken as 3 nm in the region of 350-2249 nm.

As can be seen in Fig.3, the spectral information changes mainly occurred in the two wavelength ranges of 350 nm-675 nm and 1207 nm-2249 nm. Autocorrelation peaks appeared at (467 nm, 467 nm), (674 nm, 674 nm), (1724 nm, 1724 nm), and (2144 nm, 2144 nm) in the main diagonal line, indicating that the relative intensity of these peaks changes significantly with adulteration. Positive cross-peaks appeared at (467 nm, 1724 nm), (674 nm, 1724 nm), (467 nm, 1208 nm), (467 nm, 1415 nm), (467 nm, 2144 nm), (1208 nm, 2144 nm), (1415 nm, 2144 nm) on either side of the main diagonal line, indicating that the spectral information at the autocorrelation peaks may change synergistically due to adulteration.

Two-dimensional correlation spectra can provide detailed spectral information caused by adulteration, but may also amplify the noise and reduce the signal-to-noise ratio of the spectra at the same time. As can be seen in Figure 3, the spectrum had relatively strong burr noise and low smoothness. To weaken the influence of noise interference and fully mine the effective spectral information for modeling, based on the multi-scale characteristics of spectra, BEMD algorithm was used to decompose the two-dimensional correlation synchronization spectra, and then each scale component was weighted reconstructed according to the performance of the sub-model built by the corresponding component.

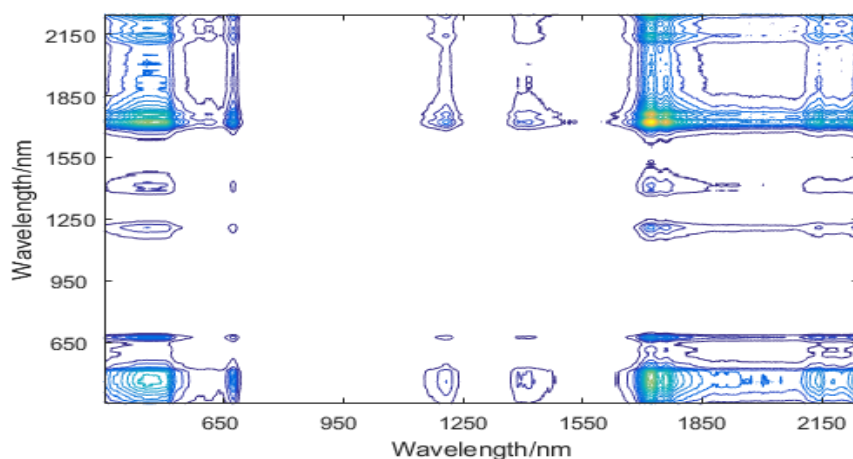


Fig. 3 - Two-dimensional correlation spectra of adulterated flaxseed oil

Weighted reconstruction of two-dimensional correlation spectra

Using the BEMD algorithm to adaptively decompose the two-dimensional correlation spectrum of each sample, six components of different scales were obtained after decomposition. Fig.4 showed the decomposed six components of the two-dimensional correlation spectrum in Fig.3. The frequencies of component 1-component 6 varied from high to low.

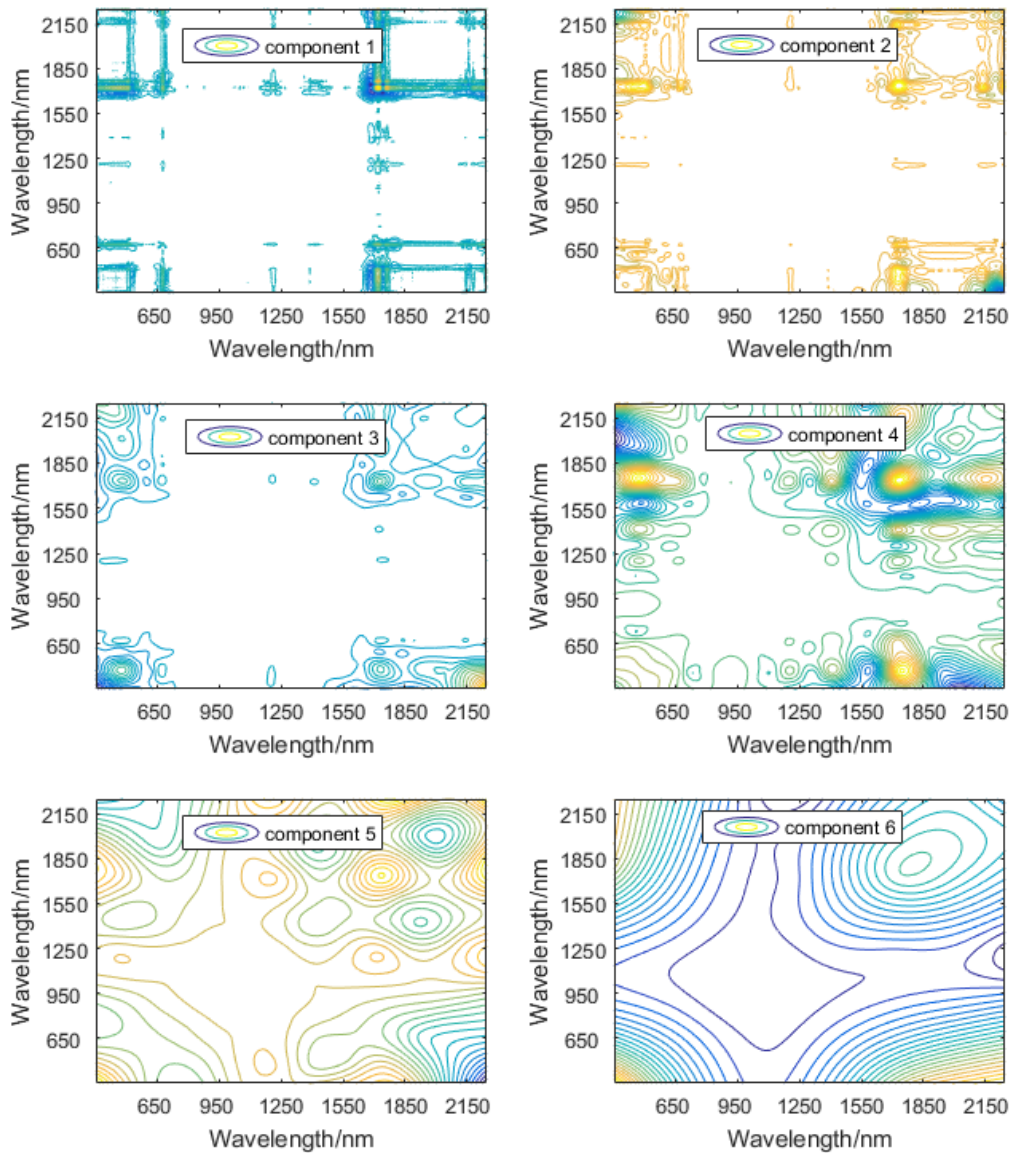


Fig. 4 - Each decomposed component of the two-dimensional correlation spectra

Based on the six components of different scales, the corresponding sub-models for flaxseed oil adulteration detection were established, and the *RMSE* values of each sub-model were calculated in terms of Eq. (2) shown in Fig. 5.

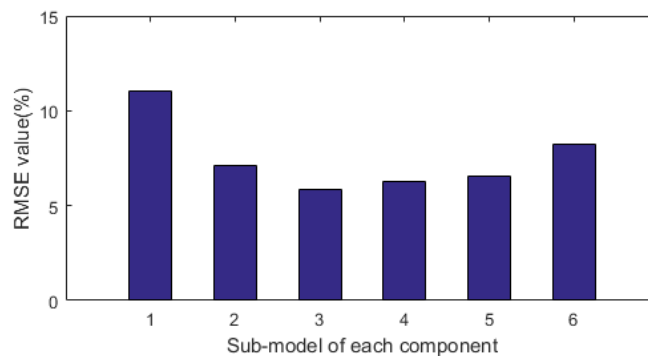


Fig. 5 - RMSE value of each sub-model based on the corresponding component

As can be seen in Figure 5, the RMSE values of the sub-models built by high-frequency component 1 and low-frequency component 6 are larger than those of the other components, because component 1 mainly contains high-frequency noise, while component 6 contains background signals. The RMSE values of the sub-models built by components 2, 3, 4, 5 are relatively smaller because these intermediate-frequency components contain more spectral information, which is in agreement with the multi-scale characteristics of spectra.

The reconstruction weight of each component was calculated in terms of Eq. (3) and shown in Fig. 6. It can be seen that the weights assigned to component 1 and component 6 were relatively smaller, and the weights assigned to component 2, component 3, component 4 and component 5 were relatively larger. The weight reassignment enhanced the intermediate-frequency components and their contribution to the modeling, reduced the proportion of high-frequency and low-frequency components, and hence suppressed spectral noise interference.

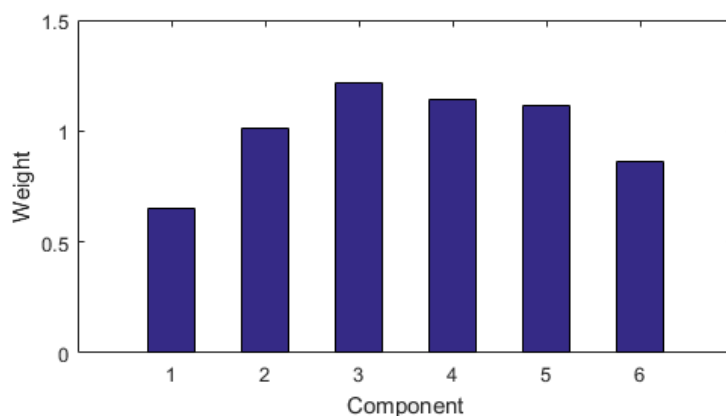


Fig. 6 - The weight assigned to each component

The two-dimensional correlation spectra of all samples were reconstructed by accumulating the weighted components and displayed in Fig.7. It can be seen that the weighted reconstructed two-dimensional correlation spectrum became clearer and smoother, and the burr noise was effectively suppressed. The weak peaks appeared at 1207 nm, 1412 nm in the main diagonal line, indicating that the relative intensity of these peaks was enhanced. The cross-peaks are well separated from the autocorrelation peaks, which improves the resolution of the spectrum.

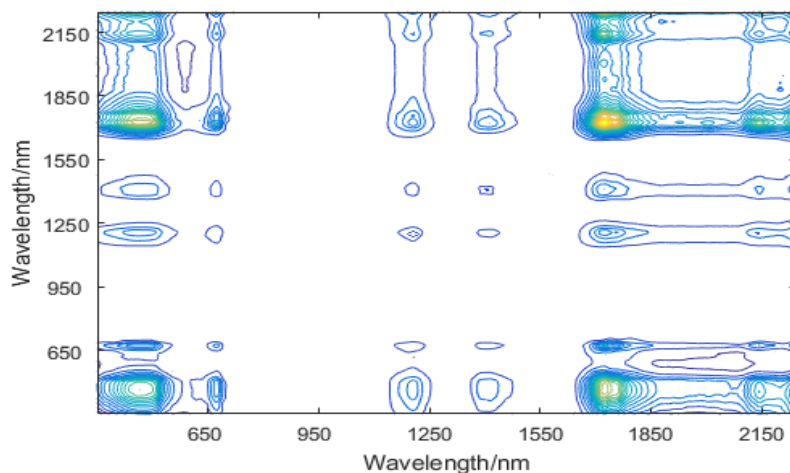


Fig. 7 - The weighted reconstructed two-dimensional correlation spectra

N-PLS model for the detection of adulterated flaxseed oil

The quantitative detection model of flaxseed oil adulteration was established using the weighted reconstructed two-dimensional correlation spectra combined with the N-PLS algorithm. The model's calibration determination coefficient (R^2_c) is 0.9375, and the root mean square error of calibration ($RMSEC$) is 2.8837%; the prediction determination coefficient (R^2_p) is 0.9252, and the root mean square error of prediction ($RMSEP$) is 2.9956%. Fig.8 and Fig.9 showed the model scatter plots of the calibration set and prediction set, respectively. The plots illustrated that the predicted adulterating ratio and actual adulterating ratio were in good agreement and the model provided a good detection precision.

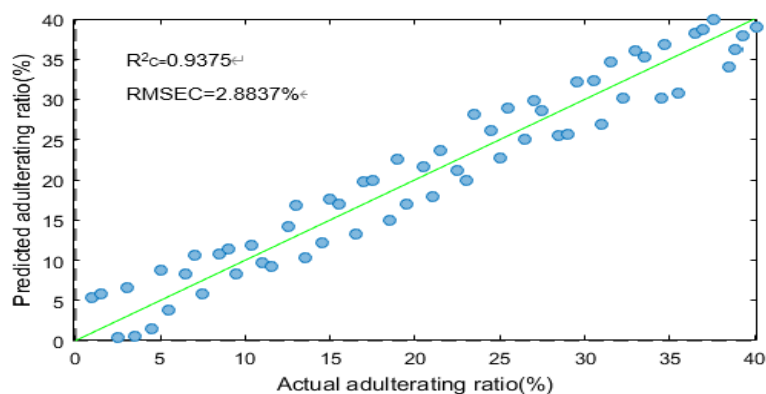


Fig. 8 - The model scatter plot of the calibration set



Fig. 9 - The model scatter plot of the prediction set

The average cutting speed and blade. To compare with the traditional two-dimensional correlation spectra (shown in figure 3), the detection model was also built based on the traditional two-dimensional correlation spectra combined with the N-PLS algorithm, and the model results are shown in Table 2.

Table 2

Results of detection model				
Item	R^2_c	R^2_p	RMSEC	RMSEP
Traditional two-dimensional correlation spectra	0.8840	0.8606	3.9283%	4.0893%
Weighted reconstructed two-dimensional correlation spectra	0.9375	0.9252	2.8837%	2.9956%

From this table, it is clear that better performance of the model can be obtained using weighted reconstructed two-dimensional correlation spectra, with the R^2_c increased by 6.05% and R^2_p increased by 7.5%, which showed that the detection model based on weighted reconstructed two-dimensional correlation spectra has higher accuracy and better prediction stability.

CONCLUSIONS

In this paper, a method was proposed for the detection of flaxseed oil adulteration based on weighted reconstructed two-dimensional correlation near-infrared spectroscopy. Based on the multi-scale characteristics of the spectra, the two-dimensional correlation near-infrared of the adulterated flaxseed oil samples were decomposed at multiple scales and weighted reconstructed. The quantitative detection model based on the reconstructed two-dimensional correlation spectra can provide better performance than the model based on the traditional two-dimensional correlation spectra with the R^2_c increased by 6.05% and R^2_p increased by 7.5%. The results showed that the weighted reconstructed two-dimensional correlation spectra can effectively reduce the spectral noise interference, enhance the spectral feature information, thereby improving the accuracy and robustness of the adulteration detection model. The proposed method explored a new perspective for the detection of adulterated oil based on two-dimensional correlation near-infrared spectra and it can also be applied to other food adulteration detection areas.

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