# NJC



### PAPER



Cite this: New J. Chem., 2023, 47, 6889

### Optimized identification of cheese products based on Raman spectroscopy and an extreme learning machine<sup>†</sup>

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Rapid and intelligent identification of similar samples is an important technical aid for food quality and safety assessment. At present, relevant research studies are still relatively scarce. In this paper, Raman spectra of different brands of cheese products with different integration times were collected and analyzed statistically. The research showed that the original Raman spectral data of different brands of cheese products had a high similarity. The conventional statistical control chart method revealed that the samples of the same brand showed certain guality fluctuations, but it was still difficult to achieve effective differentiation from other brand samples. Different spectral preprocessing methods had a greater impact on the classification accuracy of the intelligent extreme learning machine algorithm, and different spectral integration times also had a greater impact. Before optimization, the intelligent recognition algorithm had only about 40% average accuracy with the integration time of 10 s. Under the optimal conditions - an 80 s Raman spectral integration time, wavelet denoising (coif wavelet basis, 5 decomposition layers), normalization processing ([-1, 1] interval), principal component dimensionality reduction (74 principal components were extracted, which can represent 100% of the information of the original data) and 400 hidden-layer neurons - the average recognition accuracy was 98%, and the algorithm operation time was less than 5 s. The establishment of this method provides a technical reference for rapid discrimination in food quality control.

Received 30th December 2022, Accepted 13th March 2023

DOI: 10.1039/d2nj06350f

rsc.li/njc

### 1 Introduction

As a necessity of people's lives, the quality of food cannot be ignored. The common quality and safety risks mainly come from illegal additives, microorganisms and heavy-metal pollution.<sup>1,2</sup> Therefore, the methods of identification and detection are mainly component analysis, for example, quantitative determination and analysis of sodium thiocyanate in milk by surface-enhanced Raman spectroscopy.<sup>3–5</sup> Such methods can effectively realize the qualitative and quantitative analysis of specific components, but also face the limitations of cumbersome and time-consuming preprossessing. In recent years, there has been a new phenomenon in food quality risks: some cheap products have been misrepresented as high-quality food.<sup>6</sup> However, according to the analysis of specific components in-line with national standards, further relevant research

has found that their nutritional components and pollutant limits meet the requirements of food quality and safety regulations, and they are acceptable products. It is difficult for a single component index to allow an accurate judgment when detecting such quality problems.<sup>7</sup>

As a spectral technology to characterize the vibrational state of molecules, Raman spectroscopy has the advantages of direct collection of food sample signals and fast sampling speed. Further combined with chemometric algorithms, Raman spectroscopy has become a new research field in solving the problem of food quality identification.<sup>8</sup> Huang *et al.* used Raman spectroscopy combined with chemometric methods to analyze the acidity and adulteration of milk.<sup>9</sup> However, there are still relatively few studies discussing the applicable relationship between data processing and algorithm parameters, which is a key step for the effective discrimination and application of food quality identification algorithms.<sup>10</sup>

Herein, we attempt to establish a cheese product identification method based on Raman spectroscopy combined with an extreme learning machine algorithm, and optimize the spectral acquisition parameters. The present results can be summarized as follows. Firstly, the Raman spectra of cheese products of different brands are similar, and the combination

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<sup>†</sup> Electronic supplementary information (ESI) available. See DOI: https://doi.org/ 10.1039/d2nj06350f

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of spectra and an extreme learning machine algorithm can achieve efficient discrimination without traditional component analysis. Secondly, the experiment investigates the influence of Raman spectral data on the identification algorithm under different integration time conditions, and reveals that an appropriate spectral signal is conducive to improving the accuracy of the identification algorithm, which could provide guidance for the practical popularization and use of the algorithm. Finally, the identification method established in this work is accurate under the optimized conditions and has a variety of advantages. In particular, a single signal acquisition takes only 80 s, there is no sample preprossessing, it can be collected online, and the identification time of the algorithm operation is less than 5 s.

### 2 Materials and methods

Sherlock cheese products (designated as brand 1) were obtained from UK Jaredcheese Foods Co., Ltd (the United Kingdom), Miao Kelan Duo cheese products (designated as brand 2) were obtained from Shanghai Zhiran Dairy Technology Co., Ltd (Shanghai, China), and Yili cheese products (designated as brand 3) were obtained from Inner Mongolia Yili Industrial Group Co., Ltd (Inner Mongolia, China). There were 25 samples of each brand.

An appropriate amount of sample was added into each well of a 96-well plate (Corning Inc., Corning, NY, USA), and then the Raman spectrum of each cheese sample was obtained using a portable laser Raman spectrometer (ProTT-EZRaman-D3, Enwave Optronics, Irvine, CA, USA). The acquisition parameters included a laser wavelength of 785 nm, laser power of 450 mW, integration time of 10–100 s with a 10 s interval, charge-coupled device temperature of -85 °C and spectral range of 250– 2000 cm<sup>-1</sup> with a resolution of 1 cm<sup>-1</sup>. The laser output probe of the Raman spectrometer had a spot size of 100 µm in diameter. Raman spectra were collected directly without any physical or chemical pretreatment.

The baseline calibration of the obtained Raman spectra was carried out using the software SLSR Reader V8.3.9 (Enwave Optronics Inc., USA). Raman spectral data processing and analysis included wavelet denoising ("wden" function), normalization ("mapminmax" function), principal component analysis ("pca" function), an extreme learning machine ("elmtrain" and "elmpredict" function), which were carried out using the MATLAB platform (MathWorks, Natick, MA, USA). The computer configuration was as follows: central processing unit (CPU) Intel(R) Core(TM) i5-8250U CPU@1.60 GHz 1.80 GHz, and random access memory 24.0 GB.

#### 3 Results and discussion

## 3.1 Characterization and analysis of cheese products by Raman spectroscopy

The cheese product in the experimental study is a kind of reconstituted cheese containing water. Their Raman spectral



Fig. 1 Raman spectra of cheese products of (a) brand 1, (b) brand 2 and (c) brand 3 with different integration times (10-100 s).

signals can be directly collected using a spectrometer. The Raman spectra obtained in the experiment with different laser integration times for different brands of cheese products are

shown in Fig. 1. According to the existing relevant literature reports, the Raman spectra of cheese products can be assigned as shown in Table 1.<sup>11-14</sup> For example, the spectral peak at about 1760  $\text{cm}^{-1}$  is mainly from the ester C=O stretching vibration of fatty acids, while the peak that appears at about 1460 cm<sup>-1</sup> is mainly due to the deformation vibration modes of the CH<sub>2</sub> groups of fats and carbohydrates. Another particular peak appears at about 1020 cm<sup>-1</sup>, mainly from the ring breathing vibration of phenylalanine.<sup>15</sup> Since the main components of the different brands of cheese products are fat, sugar and protein, their Raman spectra are very similar, and the positions of each spectral peak are also similar, so it is very difficult to distinguish them only by the naked eve. In addition, when the integration time is 10 s, the Raman spectra of the cheese products have only a few peaks, such as those at about 1450 cm<sup>-1</sup> and 1310 cm<sup>-1</sup>. With the increase in integration time, the number of spectral peaks starts to increase, and the intensity of the spectral peaks starts to increase significantly.

## 3.2 Statistical analysis of Raman spectral fluctuation of cheese products

In the experiment, 25 samples were randomly collected from the different brands, and Euclidean distance measurements and quality control charts were used to statistically analyze their spectral changes.<sup>16,17</sup> Taking the Raman spectral data analysis of brand 1 as an example, first, the average value of its spectra was calculated, followed by the Euclidean distance between each Raman spectrum and this average value. Then, the calculation results were substituted into the calculations of the individual and moving range control charts to obtain the respective results (Fig. 2 and Fig. S1-S9, ESI<sup>+</sup>). The data in Fig. 2 are calculated and plotted based on the Raman spectral data obtained at the integration time of 80 s. Fig. 2(a and d) show the fluctuation of the individual and moving range, respectively, of each sample of brand 1 around the average value. It can be seen that two data points for the individual value have exceeded the upper control limit (UCL), one data point in the moving range has exceeded the UCL, and the other data points have fluctuated within the control limit (CL), which shows that there is a certain quality difference between the samples of brand 1,

Table 1 The main peak assignments of the Raman spectra of the different brands of cheese products<sup>11-14</sup>

but overall the data points remain within the control limits. Fig. S1-S9 (ESI<sup>†</sup>) are calculated and plotted based on the Raman spectral data obtained with different integration times, and the results are similar to those shown in Fig. 2. Fig. 2(b and e) are the individual and moving range control charts of brand 2, respectively, corresponding to the Euclidean distance results between the Raman spectral data of each sample of brand 2 and the average value of brand 1. It can be seen that only five data points have exceeded the UCL, indicating that most samples of brand 2 are similar to those of brand 1. Fig. 2(c and f) are the individual and moving range control charts of brand 3, respectively, corresponding to the Euclidean distance results between the Raman spectral data of each sample of brand 3 and the average value of brand 1. The results are also similar to those above, with only three data points exceeding the UCL, indicating that most samples of brand 3 are similar to those of brand 1. It can be seen that the quality of samples of the same brand fluctuates, and samples of different brands also have high similarity. It is difficult to achieve an ideal identification effect only using raw Raman spectral data combined with conventional descriptive statistical analysis.

### 3.3 Pretreatment analysis of Raman spectra of cheese products

In order to realize the classification and discrimination of the similar samples mentioned above, an extreme learning machine, as a chemometric method, was introduced into this research work. This method has the advantages of fast learning speed and good generalization performance.<sup>18-20</sup> Taking the Raman spectra of the samples of various brands collected with the integration time of 80 s as an example, the original data were first introduced into an extreme learning machine algorithm, with 72% of the samples as test sets (54 samples) and 28% as training sets (21 samples). The sample selection was random and ran 200 times. The number of neurons in the hidden layer of the extreme learning machine was 250. The selection of the test set and training set was random. The recognition accuracy rate was only 45%, indicating that the ideal discrimination effect was not achieved without optimization. There was a certain amount of random noise in the

Raman shift (cm <sup>-1</sup> ) – brand 1	Raman shift (cm <sup>-1</sup> ) – brand 2	Raman shift (cm <sup>-1</sup> ) – brand 3	Assignment	Possible component attribution
1758	1760	1758	$\nu$ (C=O) ester	Fat
1668	1671	1668	$\nu(C=O)$ amide I	Protein
1668	1671	1668	$\nu(C = O); \nu(C = C)$	Fat
1570	1572	1576	$\delta(N-H)$ ; $\nu(C-N)$ amide II	Protein
1457	1456	1456	$\delta(CH_2)$	Fat
1457	1456	1456	$\delta(CH_2)$	Carbohydrate
1313	1314	1314	$\tau(CH_2)$	Fat
1143	1146	1143	$\nu$ (C–O) + $\nu$ (C–C) + $\delta$ (C–O–H)	Carbohydrate
1079	1080	1080	$\nu(C-O) + \nu(C-C) + \delta(C-O-H)$	Carbohydrate
1018	1019	1019	Ring-breathing (phenylalanine); $\nu$ (C–C) ring	Protein
856	853	856	$\delta(C-C-H) + \delta(C-O-C)$	Carbohydrate

 $\nu$ : stretching vibration;  $\delta$ : deformation vibration;  $\tau$ : twisting vibration.



**Fig. 2** Quality fluctuation: individual values of brand 1 (a), brand 2 (b), and brand 3 (c) and moving ranges of brand 1 (d), brand 2 (e), and brand 3 (f) based on calculation results of the Raman spectral Euclidean distance (integration time: 80 s). UCL = upper control limit; LCL = lower control limit;  $\overline{MR}$  = the average value of the moving range control chart.  $\bar{x}$  = the average value of the individual control chart.

collected Raman spectral signal, which has a negative impact on the recognition effect of the discriminant algorithm. Therefore, the wavelet soft-threshold denoising method was applied to filter this noise. The coif1 wavelet base was used in the experiment, and the number of decomposition layers was five.<sup>21,22</sup> The results are shown in Fig. S10 (ESI†). It can be clearly seen that the Raman spectrum becomes smoother after wavelet denoising. Under the same test conditions, the recognition accuracy rate after wavelet denoising was 62%. In the process of spectrum collection, the spectral intensity value had a certain fluctuation range. In order to eliminate the influence of dimensions, the spectral intensity value was normalized to [-1, 1] using the normalization method (Fig. S11, ESI<sup>†</sup>). Under the same test conditions, the recognition accuracy rate was improved to 67%. In order to further improve the calculation efficiency, the dimensions of the Raman spectral data were reduced by principal component analysis.<sup>23</sup> Each sample in the original Raman spectrum contains 1751 data points. After the dimension reduction by principal component analysis, just 74 extracted principal components can represent 100% of the information in the original data. The cumulative interpretation Pareto diagram of the first 10 principal components is shown in Fig. S12 (ESI<sup>+</sup>), up to 77%. The first principal component can replace 40% of the original information, the second principal component can replace 11% of the original information, and the third principal component can replace 9% of the original information, as shown in Fig. 3. It can be seen from the figure that the samples of the same brand tend to cluster, but there is still some intersection between the samples of different brands in the three dimensional space. Under the same test conditions, the recognition accuracy rate was greatly



Fig. 3 Three-dimensional scatter plot of cheese products based on the results of principal component analysis.

improved to 98%. The results show that the impact of principal component dimension reduction on the classification algorithm was huge.

### 3.4 Further optimization analysis of Raman spectral identification data

The influence of the Raman spectra of cheese products with different integration times on the recognition algorithm was further studied (Fig. 4). The test condition was to randomly select 72 samples as the test set, and the remaining samples as the training set, and perform 200 random operations, with 400 neurons in the hidden layer of the extreme learning machine. Fig. 4a shows the recognition results based on the original



**Fig. 4** The recognition results based on (a) the original spectrum, (b) wavelet denoising, (c) normalization, (d) principal component analysis, (e) wavelet denoising combined with normalization, (f) normalization combined with principal component analysis, and (g) wavelet denoising, normalization and principal component analysis.

Raman spectral data. It can be seen that the different integration times correspond to different recognition accuracy rates. With the increase in integration time, the recognition accuracy rate shows an upward trend. The lowest value is 40% at the integration time of 10 s, and the highest value is 61% at the integration time of 80 s, which shows that it is difficult to realize effective recognition under such conditions. After wavelet denoising, the average recognition accuracy changes insignificantly under these conditions, and even shows a downward trend at an integration time of 50 s (Fig. 4b). This phenomenon is different from the situation in Fig. S13 (ESI<sup>†</sup>). The number of neurons in the hidden layer of the extreme learning machine in Fig. S13 (ESI<sup>+</sup>) is 250. The average recognition accuracy after wavelet denoising shows an obvious upward trend after an integration time of 40 s. This result shows that the effect of wavelet denoising is closely related to the number of neurons in the hidden layer of the extreme learning machine. The normalized processing result is shown in Fig. 4c. The average recognition accuracy shows an upward trend with the increase in integration time, which is close to the recognition result in Fig. 4a, except after 80 s. Fig. 4d shows the processing results of principal component analysis. The average recognition rate shows a good upward trend, and the maximum value is 85% at 100 s. Fig. 4e and f show the average recognition accuracy obtained by combining wavelet denoising and normalization processing, and normalization and principal component analysis, respectively. It can be clearly seen that the average recognition accuracy increases significantly after combining processing methods. The maximum value in Fig. 4e is 81%, corresponding to the integration time of 100 s, and the maximum value in Fig. 4f is 97%, corresponding to the integration time of 80 s. When wavelet denoising, normalization and principal component analysis are combined, with the results shown in Fig. 4g, the average recognition accuracy reaches 98%



**Fig. 5** Analysis of the influence of the number of hidden-layer neurons on the performance of the extreme learning machine.

after integration time of 80 s. This result shows that the combined pretreatment technology has the effect of jointly increasing the average recognition accuracy compared with a single processing method.

In the extreme learning machine algorithm, the number of neurons in the hidden layer is also an important factor. Research has been carried out for different integration times and different numbers of neurons in the hidden layer. Spectral processing includes wavelet denoising, normalization and principal component analysis, which are also used to process spectral data. The results are shown in Fig. 5, which shows that the number of neurons in different hidden layers has certain effects with different integration times. In the 50–200 range of the number of neurons, the overall average recognition accuracy rate shows an upward trend with the increase in the number of neurons. When the number of neurons reaches 250,



**Fig. 6** Recognition results of cheese products based on the extreme learning machine (*y*-axis: label 1 represents brand 1, label 2 represents brand 2, and label 3 represents brand 3).

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the recognition accuracy rate increases slowly and tends to be stable. When the number reaches 400, the recognition accuracy rate basically changes little. Fig. 6 shows the recognition results of cheese products based on the optimized conditions. Based on the above research, under the optimized experimental conditions, namely an 80 s Raman spectral integration time of cheese products, wavelet denoising, normalization processing, principal component dimensionality reduction, and 400 hidden-layer neurons, the average recognition accuracy of the recognition algorithm can reach 98%. The operation time of this algorithm is less than 5 s.

### 4 Conclusions

For cheese products with high similarity, Raman spectra can be collected directly, but the spectra have high similarity. Conventional statistical analysis shows that the samples have a certain quality fluctuation, but classification has not been realized. Further research shows that the integration time of Raman spectroscopy and preprocessing of the spectral data have a great impact on the recognition effect of the extreme learning machine algorithm. After optimization, under the conditions of 80 s integration time, wavelet denoising, normalization, principal component dimension reduction, and 400 hidden-layer neurons, the average recognition accuracy can reach 98%, and the recognition algorithm operation time is less than 5 s. This scheme has the advantages of convenient signal acquisition and fast calculation speed, which provides a new research route for efficient discrimination of similar samples.

### Author contributions

The overall work was organized, drafted and finally written by Zheng-Yong Zhang and Huan-Ming Xiong. The experiment and data analysis was performed by Min-Qin Jiang and Zheng-Yong Zhang. All authors have given approval to the final version of the manuscript.

### Conflicts of interest

There are no conflicts to declare.

### Acknowledgements

This research was financially supported by the Excellent Young Backbone Teachers of "Blue Project" in Jiangsu Universities in 2021, the Industry University Research Collaboration Foundation of Jiangsu Province (BY2022611), and the National Natural Science Foundation of China (61602217).

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