Raman chemical feature extraction for quality control of dairy products

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ABSTRACT

As a fast information acquisition technique, Raman spectroscopy can be used to control the quality of dairy products. Feature extraction is a necessary processing step to improve the efficiency of Raman spectral data. Principal component analysis is a traditional method that can effectively extract the features and reduce the dimension of spectral data. However, it is difficult to analyze the chemical information of the extracted feature, thus limiting its practical application. In this work, Raman spectral chemical feature extraction was carried out. The quality control of Dingxin dairy products (a famous dairy brand in China; purchased from Heilongjiang Zhaodong Tianlong Dairy Co. Ltd., Heilongjiang, China) was used as an example. Raman peak intensity, peak area, and peak ratio were extracted as chemical features and further investigated using Euclidean distance and the quality fluctuation control chart. The potential quality discrimination ability of the Raman feature extraction methods was demonstrated. The results showed that the Puzhen dairy products (purchased from Inner Mongolia Yinuo Halal Food Co. Ltd., Inner Mongolia, China) and Xueyuan dairy products (used as a control; purchased from Inner Mongolia Wulanchabu City Jining Xueyuan Dairy Co. Ltd., Inner Mongolia, China) could be distinguished from Dingxin dairy products when the Raman chemical features special peak intensity, peak area, and peak ratio were used, and their discriminatory ability increased in sequence. Hence, it was shown that Raman chemical feature extraction can achieve quality control and discriminant analysis of dairy products and that the spectral information is clear.

Key words: feature extraction, Raman spectroscopy, dairy product, quality control

INTRODUCTION

The quality and safety control of dairy products is a hot issue for regulators, manufacturers, and consumers. The traditional quality control methods of dairy products can be attributed to the following 2 strategies. The first strategy is the sensory evaluation method, which focuses on people's feelings, but its disadvantage is that people's subjective evaluation may be affected by the environment and their health status. The second strategy is instrumental analysis, which includes the separation analysis method (e.g., chromatography), the bionic identification method (e.g., electronic nose), and the fast analysis method (e.g., spectroscopy; Peris and Escuder-Gilabert, 2016; Chen et al., 2017; Poonia et al., 2017). The chromatographic separation method can separate various components of the sample and make qualitative and quantitative analysis according to the standard curve method. The bionic identification method mainly uses sensors to simulate the biological response for the test molecules and then combined the chemometrics model to analyze the quality of samples. However, these methods are often time consuming (Nieuwoudt et al., 2016a). The fast analysis method, especially Raman spectroscopy, has the advantages of not needing complicated sample pretreatment process, fast scanning speed, high efficiency, and on-line and nondestructive testing and has been developed rapidly in recent years (Nieuwoudt et al., 2016b).

Raman spectroscopy can provide rich molecular vibration information about the sample, which can be used for qualitative, quantitative, and structural analysis of characteristic molecules. There are 2 main ways to control the quality of dairy products by Raman spectroscopy. One is direct analysis of characteristic molecular concentration based on the Beer-Lambert law. A standard curve can be constructed according

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to the relationship between the characteristic molecule content and the peak intensity or the peak area of Raman spectroscopy. For example, we have used the standard curve method to quantitatively analyze the content of sodium thiocyanate (an illegal additive) in dairy products (Zhang et al., 2015). This method can provide the content information of the Raman active substances, but the data processing usually simply uses the information of specific peaks, and the data utilization rate is relatively low. The other is the quality discriminant analysis based on the chemometrics method, such as artificial neural network and partial least squares (Alves da Rocha et al., 2015; Wang et al., 2015; Mendes et al., 2016). Principal component analysis is a common feature extraction algorithm that can reduce the interference of redundant information and improve the efficiency of the discriminant algorithm. However, principal component analysis extraction is a mathematical transformation method. It is difficult to analyze the chemical information of the extracted features, which limits its migration and popularization (Cebi et al., 2017). Therefore, developing more practical quality control methods that fully use Raman chemical information for dairy products is a hot topic.

Herein, we have developed a quality control method for dairy products based on Raman chemical feature extraction, which mainly includes the following 3 aspects. First, the traditional quality control methods for dairy products are mainly based on the analysis of the content of the nutritional ingredients or illegal additives, so it is almost impossible to make effective brand discrimination of dairy products (Qi et al., 2016; Rodrigues Júnior et al., 2016). In 2016, fake and shoddy dairy products were falsely presented as high-quality dairy products in Shanghai, and the main ingredients of these products were in conformity with the national standards for the quality and safety of dairy products (Zhang et al., 2017a). The method presented has great potential application for brand discriminant analysis of dairy products. Second, the discriminant analysis of traditional dairy products is based on relatively complex feature extraction. For instance, principal component analysis is a pure mathematical transformation. This study is based on the analysis of chemical characteristics, which will be simpler than the principal component analysis and extend the direct application of Raman spectroscopy. Finally, chemometric algorithms for traditional discriminant analysis of dairy products are relatively complex and have the risk of overfitting. A statistical control chart has been applied in this study, which is more intuitive and suitable for the actual quality control application.

MATERIALS AND METHODS

Samples and Instruments

Dingxin (**DX**) dairy products were purchased from Heilongjiang Zhaodong Tianlong Dairy Co. Ltd. (Heilongjiang, China). Puzhen (**PZ**) dairy products were purchased from Inner Mongolia Yinuo Halal Food Co. Ltd. (Inner Mongolia, China). Xueyuan (**XY**) dairy products were purchased from Inner Mongolia Wulanchabu City Jining Xueyuan Dairy Co. Ltd. (Inner Mongolia, China). They were all milk products. There were 49 samples of DX dairy products, 42 samples of PZ dairy products, and 60 samples of XY dairy products.

The Raman spectra were collected from a portable laser Raman spectrometer; then, baseline calibration of the recorded Raman spectra was carried out by the software SLSR Reader V8.3.9, Prott-ezRaman-d3 (Enwave Optronics, Irvine, CA). The excitation wavelength of the laser was 785 nm, the laser power was 450 mW, and the integration time was 50 s. The spectrometer operated from 250 to 2,339 cm^{-1} with a resolution of 1 cm^{-1} .

Data Analysis

Euclidean Distance. Euclidean distance is a method commonly used to measure distance. The formula for calculating the Euclidean distance between 2 samples in the n dimensional space is

$$d = \sqrt{\sum_{i=1}^n (x_i - y_i)^2}.$$

In the formula, d represents the Euclidean distance, and x_i and y_i represent the i dimensional components of the x and y samples, respectively. In this work, x_i and y_i represent the spectral intensity (or area; ratio) of 2 samples at a specific band; i represents the spectral wavenumber.

Statistical Control Chart. The control chart can be prepared using the following individual and moving range chart formulae. For the individual (d) control chart, the formula is as follows:

$$\begin{cases} UCL_d = \bar{d} + 2.66\bar{R} \\ CL_d = \bar{d} \\ LCL_d = \bar{d} - 2.66\bar{R} \end{cases}.$$

For the moving range (MR) control chart, the formula is as follows:

$$\begin{cases} UCL_{MR} = 3.267\bar{R} \\ CL_{MR} = \bar{R} \\ LCL_{MR} = 0 \end{cases}.$$

In the formulae, d and $\bar{d}$ represent the Euclidean distance and the average value of the samples, respectively; R represents the moving range, which is $R = |d_{i+1} - d_i|$; d_i represents the Euclidean distance of the sample i variable; $\bar{R}$ represents the average value of R ; CL represents the center line; UCL represents the upper control limit; and LCL represents the lower control limit.

Calculation of Characteristic Peak Area of Raman Spectroscopy. According to the trapezoidal method, the feature peak area is calculated as follows:

$$\int_a^b f(x)dx = \frac{b-a}{2N} \sum_{n=1}^N [f(x_n) + f(x_{n+1})].$$

In the formula, $f(x)$ represents the trapezoid function, a represents the number of the initial band of the feature peak, b represents the number of the end band of the feature peak, N represents the number of the interval between the initial band and the end band of the feature peak ($N = b - a$), and n represents the spectral wavenumber of the a and b segments. The computing platform used was Matlab R2013a (MathWorks, Natick, MA).

RESULTS AND DISCUSSION

Raman Spectroscopic Analysis of Dairy Products

The Raman spectra of dairy products are shown in Figure 1. The main vibrational bands are assigned as follows according to the previous reports (McGoverin et al., 2010; Almeida et al., 2011; Mazurek et al., 2015; Júnior et al., 2016; Zhang et al., 2017c). The Raman band at $1,752 \text{ cm}^{-1}$ could be attributed to the C=O stretching ester of fatty acids. The band at $1,660 \text{ cm}^{-1}$ could be associated with the C=O stretching from the CONH group of amide I of proteins and C=C stretching mode from UFA. The band at $1,462 \text{ cm}^{-1}$ could be a contribution from CH_2 deformation of fats and carbohydrate molecules. The band at $1,337 \text{ cm}^{-1}$ could be assigned to C–O stretching and C–O–H deformation of carbohydrate molecules. The band at $1,307 \text{ cm}^{-1}$ could be attributed to CH_2 twisting of lipid molecules. The band at $1,256 \text{ cm}^{-1}$ could be attributed to CH_2 twisting of carbohydrate molecules. There are about 6 bands in the range of 800 to $1,200 \text{ cm}^{-1}$, which were attributed

to carbohydrates, such as C–C stretching and C–O–H deformation ($1,130 \text{ cm}^{-1}$, $1,078 \text{ cm}^{-1}$, and $1,027 \text{ cm}^{-1}$), C–O–C deformation (922 cm^{-1} and 861 cm^{-1}), and the ring breathing of phenylalanine of protein ($1,007 \text{ cm}^{-1}$, the band could be attributed to C–C symmetric stretching in rings of phenylalanine, with phenylalanine from protein). There are about 7 bands in the range of 250 to 800 cm^{-1} , which could be attributed to C–C–O deformation (777 cm^{-1}), C–S stretching (718 cm^{-1}), C–C–C deformation and C–O twisting (570 cm^{-1}), glucose (520 cm^{-1}), C–C–C deformation and C–O twisting (487 cm^{-1}), glucose (425 cm^{-1}), and lactose (373 cm^{-1}).

Feature Extraction and Analysis of Dairy Products Based on Raman Peak Intensity

It can be seen from the above analysis that Raman spectra contain rich molecular information about dairy products. In this paper, DX dairy products (a famous dairy brand in China) were used as the main research object, and the investigation of its quality control based on the feature extraction of Raman spectra was carried out. For product quality control, there are 2 important aspects. First, the quality fluctuation of products should be within a reasonable space. Second, the other brands' products are not in the controllable range and can be distinguished from the DX dairy products. Spectral intensity is the most commonly used direct feature of Raman spectroscopy, which reflects the information of the characteristic components of the sample. In feature selection, the full spectrum or a specific band spectrum can be selected. First, the full spectra of the collected Raman spectroscopy ($250\text{--}2,339 \text{ cm}^{-1}$) were used as data inputs. For the description of sample quality, the Euclidean distance method was used in this work. The Euclidean distance algorithm can evaluate the similarity between 2 samples. When the similarity between the samples is high, the Euclidean distance is small; conversely, when the similarity between the samples is low, the Euclidean distance is large (Chen et al., 2009). The average spectrum of all the collected Raman spectra of DX dairy products was taken as the theoretical true value of the sample, and then the Euclidean distances between each experimental sample and the theoretical true value were calculated. Next, an individual and moving range quality fluctuation control chart for DX dairy products was constructed using the calculated Euclidean distances, which can evaluate the quality stability of the samples (Zhang et al., 2017b), as shown in Supplemental Figure S1 (<https://doi.org/10.3168/jds.2018-14569>). The calculated Euclidean distances fluctuate around the center line, indicating that there is a certain quality fluctuation among the

Figure 1. Raman spectra of different dairy products. DX = Dingxin dairy products purchased from Heilongjiang Zhaodong Tianlong Dairy Co. Ltd. (Heilongjiang, China). PZ = Puzhen dairy products purchased from Inner Mongolia Yinuo Halal Food Co. Ltd. (Inner Mongolia, China). XY = Xueyuan dairy products purchased from Inner Mongolia Wulanchabu City Jining Xueyuan Dairy Co. Ltd. (Inner Mongolia, China).

samples. According to the normal distribution model, the upper and lower control limits were calculated. The results show that although the Euclidean distance values of the sample fluctuate all the time, they are all controlled by the upper and lower control limits. Supplemental Figure S1 reflected a high stability among DX dairy products when the full spectra were applied as the feature inputs. The Euclidean distances (average value) among the PZ, XY (the other famous dairy brands in China; used as control), and DX (experimental group) dairy products were calculated, as shown in Supplemental Figure S2 (<https://doi.org/10.3168/jds.2018-14569>). It can be seen that 5 samples of PZ dairy products are lower than the control limit and that 2 samples of XY dairy products are near the control limit. Supplemental Figure S1 identified the quality stability of the same kind of dairy products but we had difficulty distinguishing the different dairy products (the full spectra as feature inputs). The main reason is that there is noise and redundant information in Raman spectroscopy. Therefore, it is necessary to further extract chemical features of Raman spectroscopy to improve the capability of data discrimination.

Through the direct comparison analysis of the Raman spectra, it can be found that the Raman intensity at $1,307\text{ cm}^{-1}$ among the samples is different. According to the above Euclidean distance calculation and

statistical control chart method, the quality fluctuation control chart of DX dairy products can be obtained based on the Raman intensity at $1,307\text{ cm}^{-1}$, as shown in Figure 2. We can see that the Euclidean distances among the DX dairy products decreased significantly in Supplemental Figure S1 (<https://doi.org/10.3168/jds.2018-14569>), and all samples fluctuated around the center line; no samples were out of the control limit. This result shows that the same dairy products have high similarities among themselves. The Euclidean distances (average value) among the PZ, XY, and DX dairy products were calculated by the same operation method as shown in Figure 3. It can be seen that 12 samples of PZ dairy products were dropped into the control limit, whereas XY dairy products are all separated. It is suggested that the quality discrimination ability is limited when the Raman spectral intensity is used as feature input only.

Feature Extraction and Analysis of Dairy Products Based on Raman Peak Area

The peak area of Raman spectroscopy is the other feature source of spectrogram analysis, which reflects the more abundant information of characteristic molecules of the sample. Because the peak area involves a spectral distance, it often contains the interaction

Figure 2. Quality fluctuation individual value (A) and moving range (B) control chart of Dingxin dairy products (purchased from Heilongjiang Zhaodong Tianlong Dairy Co. Ltd., Heilongjiang, China) based on Euclidean distances of Raman peak intensities (at $1,307\text{ cm}^{-1}$). UCL = upper control limit; LCL = lower control limit; MR = the average value of moving range control chart.

information of multiple characteristic molecules in the peak range. The peak areas (full spectra, $250\text{--}2,339\text{ cm}^{-1}$) were used as the feature inputs first. The Euclidean distances among the samples and DX dairy products (average value) were calculated and the quality

control chart was presented, as shown in Supplemental Figure S3 (<https://doi.org/10.3168/jds.2018-14569>). The results showed that all samples fluctuated around the center line and fell within the control limits. The Euclidean distances (average value) among the PZ, XY, and DX (experimental group) dairy products were obtained under the same calculation condition, as shown in Supplemental Figure S4 (<https://doi.org/10.3168/jds.2018-14569>). The results showed that 37 samples of PZ dairy products and 45 samples of XY dairy products fell into the control limit, indicating that the peak areas of full spectrum as inputs cannot distinguish the different samples. After spectral analysis, the peak area in the range of $1,288\text{ to }1,319\text{ cm}^{-1}$ was selected for feature input. The Euclidean distances (average value) between each sample and the DX dairy products and the control chart were calculated as previously mentioned, as shown in Figure 4. The results in Figure 4 suggest that quality fluctuations exist, and 1 sample is beyond the control limit. This reveals that there may be a quality risk, but the rest of the samples fall within the control limits, so the overall quality level of DX dairy products is still in the controllable and stable state. Figure 5 shows the discriminant analysis results (average value) of the PZ, XY, and DX dairy products under the same condition. All dairy products

Figure 3. Euclidean distances (mean value) among Puzhen dairy products (PZ; purchased from Inner Mongolia Yinuo Halal Food Co. Ltd., Inner Mongolia, China) and Xueyuan dairy products (XY; purchased from Inner Mongolia Wulanchabu City Jining Xueyuan Dairy Co. Ltd., Inner Mongolia, China) based on Raman peak intensities (at $1,307\text{ cm}^{-1}$).

Figure 4. Quality fluctuation individual value (A) and moving range (B) control chart of Dingxin dairy products (purchased from Heilongjiang Zhaodong Tianlong Dairy Co. Ltd., Heilongjiang, China) based on Euclidean distances of Raman peak areas (at 1,288 to 1,319 cm^{-1}). UCL = upper control limit; LCL = lower control limit; MR = the average value of moving range control chart.

(control group) fall outside of the control limit, and only a few samples of the PZ dairy products seem close to the control limit. The results show that peak area as a feature can provide more abundant information about samples, but the full spectrum area as input will fail to

meet the needs of quality discriminant application. A discriminant model of higher quality can be carried out through the appropriate input of feature peak area.

Feature Extraction and Analysis of Dairy Products Based on the Ratio of Raman Peaks

Peak ratio of Raman spectroscopy can be used as a new feature. For the production of dairy products, their raw material ratio and production process is similar, so the peak ratio of the same kinds of products is highly consistent. Spectral peak ratios of the intensities at 1,337 cm^{-1} /1,307 cm^{-1} were selected as feature inputs, and the Euclidean distances between each DX dairy product sample and their average value and the control chart were calculated, as shown in Figure 6. The Euclidean distances among the experimental samples fluctuate around the center line, and they all fall within the control limit, indicating that the sample has a high stability and consistency. Figure 7 shows the samples' relationship among the PZ, XY, and DX dairy products (average value). The results show that the Euclidean distance of the PZ and XY dairy products is far away from the control limit of the DX dairy products, which can effectively distinguish the dairy products (control group) from the experimental group. The reason is

Figure 5. Euclidean distances (mean value) among Puzhen dairy products (PZ; purchased from Inner Mongolia Yinuo Halal Food Co. Ltd., Inner Mongolia, China) and Xueyuan dairy products (XY; purchased from Inner Mongolia Wulanchabu City Jining Xueyuan Dairy Co. Ltd., Inner Mongolia, China) based on Raman peak areas (at 1,288 to 1,319 cm^{-1}).

Figure 6. Quality fluctuation individual value (A) and moving range (B) control chart of Dingxin dairy products (purchased from Heilongjiang Zhaodong Tianlong Dairy Co. Ltd., Heilongjiang, China) based on the Euclidean distances of the ratio of Raman peaks (at $1,337\text{ cm}^{-1}/1,307\text{ cm}^{-1}$). UCL = upper control limit; LCL = lower control limit; MR = the average value of moving range control chart.

that the ratios of the material content among the dairy products are differential, and the quality difference can be distinguished effectively through the ratio of peak to peak.

Figure 7. Euclidean distances (mean value) among Puzhen dairy products (PZ; purchased from Inner Mongolia Yinuo Halal Food Co. Ltd., Inner Mongolia, China) and Xueyuan dairy products (XY; purchased from Inner Mongolia Wulanchabu City Jining Xueyuan Dairy Co. Ltd., Inner Mongolia, China) based on the ratio of Raman peaks (at $1,337\text{ cm}^{-1}/1,307\text{ cm}^{-1}$).

Quality Discriminant Analysis of Dairy Products Based on Raman Multiple Features

This paper developed a feature extraction method based on Raman peak intensity, peak area, and peak ratio and presented the application value of experimental sample quality control from 3 dimensions. The Euclidean distances (average value) among the PZ, XY, and DX dairy products were calculated. The 3-dimensional spatial distribution map of the samples was plotted with peak intensity, peak area, and peak ratio of x, y, and z axes, respectively, as shown in Figure 8. The DX (experimental) group can be effectively separated from the control group of the PZ and XY dairy products. Therefore, a multidimensional quality control system for dairy products can be constructed successfully and can be widely applied in further practical application.

CONCLUSIONS

Raman spectroscopy is a powerful tool for rapid analysis and quality identification of dairy products because it can provide rich molecular information with rapid scanning speed. Spectral feature extraction can improve the efficiency of the discriminant method, and compared with the traditional mathematical conversion

Figure 8. A Euclidean distance 3-dimensional map of dairy products based on the peak intensity, peak area, and peak ratio of Raman spectra. DX = Dingxin dairy products purchased from Heilongjiang Zhaodong Tianlong Dairy Co. Ltd. (Heilongjiang, China). PZ = Puzhen dairy products purchased from Inner Mongolia Yinuo Halal Food Co. Ltd. (Inner Mongolia, China). XY = Xueyuan dairy products purchased from Inner Mongolia Wulanchabu City Jining Xueyuan Dairy Co. Ltd. (Inner Mongolia, China).

feature extraction method, chemical feature extraction will be more helpful for the analysis of chemical information. Raman peak intensity, peak area, and peak ratio were applied as chemical features as well as combined with Euclidean distance, and statistical control chart methods were investigated. The results show (1) that the discriminant ability using feature extraction of the special band is greater than the full spectrum as input, (2) that the spectral intensity, spectral area, and spectral ratio can be used as chemical feature inputs and that their discriminant ability increases sequentially, and (3) that these extracted chemical features can be used for quality control of dairy products.

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