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Simultaneous determination of total polyphenols and caffeine contents of green tea by near-infrared reflectance spectroscopy

Quansheng Chen [a,*], Jiewen Zhao [a,*], Xingyi Huang [a], Haidong Zhang [b], Muhua Liu [c]

[a] School of Biological and Environmental Engineering, Jiangsu University, 212013 Zhenjiang, P.R. China
[b] Faculty of Engineering and Technology, Yunnan Agricultural University, 650201 Kunming, P.R. China
[c] College of Engineering, Jiangxi Agricultural University, 330045 Nanchang, P.R. China

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Abstract

This paper indicates the possibility to use near infrared (NIR) spectroscopy as a rapid method to predict quantitatively the content of caffeine and total polyphenols in green tea. A partial least squares (PLS) algorithm is used to perform the calibration. To decide upon the number of PLS factors included in the PLS model, the model is chosen according to the lowest root mean square error of cross-validation (RMSECV) in training. The correlation coefficient R between the NIR predicted and the reference results for the test set is used as an evaluation parameter for the models. The result showed that the correlation coefficients of the prediction models were $R=0.9688$ for the caffeine and $R=0.9299$ for total polyphenols. The study demonstrates that NIR spectroscopy technology with multivariate calibration analysis can be successfully applied as a rapid method to determine the valid ingredients of tea to control industrial processes.

Keywords: Near infrared spectroscopy; PLS; Green tea; Caffeine; Total polyphenols

1. Introduction

Tea is one of the most popular beverages worldwide, which is of great interest due to its beneficial medicinal properties [1]. There is increasing evidence that specific substances found in certain foods can enhance general healthy. Recent research suggests that antioxidants found in tea may play an important role to prevent cardiovascular disease [2], chronic gastritis [3,4] and some cancers [5,6]. Moreover, an observational study in Japan found that the regular consumption of green tea (more than 3 cups a day) might be protective against recurrence of breast cancer in the early stages [7]. With the increasing consumption of the tea, quality control of tea becomes more and more important nowadays, for example, many national and international authorities are setting criteria for quality factors. In generally, total polyphenols and caffeine content are analyzed as the important tealeaves quality factors. Total polyphenols content account for more than 30% of the dry weight of tealeaves. These compounds are mainly responsible for the characteristic astringent and bitter taste of tea brews [8]. In addition, caffeine in tea, known for their stimulative effect, has to be recognized as important quality factors in tealeaves. In contrast to the catechins in polyphenols, caffeine can enhance observably tea flavor.

In the past few years, different methods of analysis had been employed to determine the contents of the compounds in question. Some approaches such as high performance liquid chromatography (HPLC) [9] and capillary electrophoresis [10] were applied to determine the caffeine content in tea. Some other approaches have also been described to estimate the content of total polyphenols using colorimetric measurements and the titration using potassium permanganate [11]. However, all of the methods mentioned above are time-consuming. Near infrared reflectance spectroscopy is a fast, accurate and non-destructive technique that can be employed as a replacement of time-consuming chemical method.

Near-infrared (NIR) spectroscopy has proved to be a powerful analytical tool for analyzing quantitative caffeine content in coffee [12–14]. Some studies on analyzing tea by

* Corresponding authors. Tel.: +86 511 8790308; fax: +86 511 8780201.
E-mail address: chenjiang0518@yahoo.com.cn (Q. Chen).

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NIR spectroscopy are reported, for example, it was used for measuring the theaflavin and moisture contents as well as for the prediction of black tea quality by Hall [15]. The prediction of quality parameters (like catechins, gallic acid, caffeine and theobromine) in green tealeaves by NIR was also reported by Schulz [16]. Recently, Luypaert and Zhang et al. [17,18] attempted the feasibility for prediction of total antioxidant capacity in green tea using NIR. Although they gave some better results for tea using NIR, they had no details in discussing the prediction models even not use an independent test set to test the robustness of the model, such as Schulz [16].

The prerequisite of NIR application for quantitative purpose is building a reliable calibration model. In this paper, our aim is to prove the applicability of multivariate calibration to NIR data. We systematically study the different steps that have to be gone through in multivariate calibration. PLS model is used and focused on the effect on the principal component factor and the method of spectra preprocessing. The robustness of the final PLS model is evaluated according to the root mean square error of cross-validation (RMSECV), the root mean square error of prediction (RMSEP) and the correlation coefficient (*R*).

2. Materials and methods

2.1. Sample preparation

All tea samples come from different provinces in China, and they have been all already on stock within 4 months period. Taking into consideration the heterogeneity of tea samples, major attention is paid to the sampling stage, and the samples would be grinded before analysis. For the grinding, the whole tealeaves are put into a small electric coffee mill and ground during 10 s. After this procedure, the powders are sieved with a mesh width 500 μm and these sieved powders are used for the further analysis.

2.2. Chemical analysis

2.2.1. High performance liquid chromatography (HPLC)

Approximately 2.0 g of the powdered material, accurately weighed, is extracted twice with 80 mL of 70% aqueous methanol each for 30 min at a temperature of 80 °C. After cooling, the extracts are centrifuged at 3000 rpm for 10 min. The liquid phases of both extracts are collected in a 250-mL volumetric flask and made up to volume by 70% aqueous methanol. The tea brew is filtered through a 0.45-μm membrane filter, diluted 5 times with Millipore water and analyzed immediately.

To determine the content of caffeine, *RP-HPLC* method is applied in the Agilent 1100 series (Aligent, USA). The used column is a deactivated monomeric ultrapure silica Zorbax Rx-C18 column with 4.6 mm × 250 mm (i.d. × length) and 5 μm nominal particle size. The flow rate is set at 1.0 ml/min and the injected volume is 50 μl. The column temperature is kept at 35 °C using a column oven. Eluents are water/acetonitrile (9:1, v/v). The caffeine of the separation is checked by its spectra recorded using the DAD and the UV-detector is set at 276 nm. The HPLC separation of the caffeine is shown in Fig. 1.

2.2.2. Colorimetric measurements [11]

Total polyphenols are estimated by a photometric Folin-Ciocalteu assay according to a proposed international standard method. Absorbance (*E*) at 540 nm of the reaction solution is determined in a 1-cm light-path cell by a ***Lengguang-752***

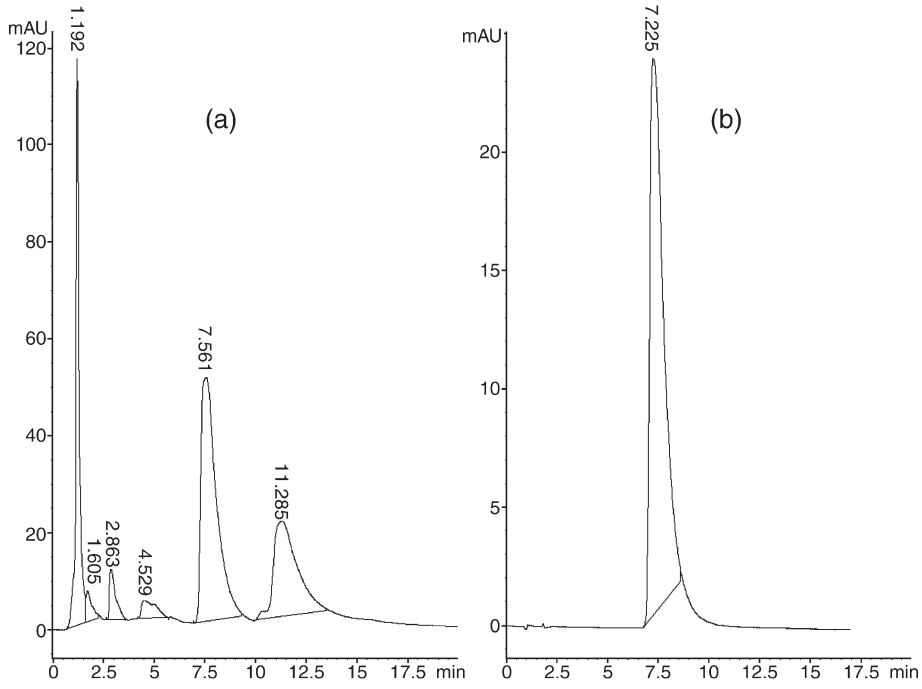

Fig. 1. HPLC separation of caffeine: (a) Chromatogram of tea samples; (b) Chromatogram of caffeine as calibration standard.