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Building models to evaluate internal comprehensive quality of apples and predict storage time

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ABSTRACT

Apple quality and customer satisfaction are significantly impacted by variations in apple quality throughout storage. In this study, the evaluation of apples' internal overall quality and the estimation of storage time were investigated. By using Pearson correlation analysis and an analytical hierarchy approach, an internal comprehensive quality index was created. A calibration model and a high-order kinetic model were created for the internal comprehensive quality index using the competitive adaptive reweighted sampling (CARS) algorithm in conjunction with partial least squares regression (PLSR) and in accordance with the results of fitting chemical kinetic reactions to variations in internal comprehensive quality with storage time. The calibration model and the high-order kinetic model were combined to create a prediction model for the storage time of apples. Results revealed that the determination coefficient of the prediction (R_p^2) and root mean square error (RMSE) of the calibration model were 0.9419 and 0.0023 respectively, and a residual predictive deviation (RPD) of 5.77; the correlation coefficient (R) and RMSE of the higher-order kinetic model were 0.9620 and 0.0038 respectively; the R_p^2 of the prediction model was determined as 0.8957, with a root mean square error of 4.63 d. Results show that the proposed calibration model and higher-order kinetic model are capable of evaluating the internal comprehensive quality of apples, and that the determined prediction model is capable of projecting the storage time of apples with an acceptable margin of error while still meeting the real requirements.

1. Introduction

Apples are liked by customers because of their high content of dietary fiber, minerals, and multivitamins, which provide them antioxidant, anti-aging, and immunity-boosting properties [1–3]. The amount of fresh food consumed when eating apples is the highest, and internal quality affects the texture and flavor of the fruit, influencing customer willingness and consumption levels [4,5]. The performance of the parameters that can reflect a specific aspect of an apple's internal quality can be relied upon by a select few consumers to satisfy their personalized consumption, but the majority of consumers prefer to select the apples that perform best overall across all internal quality-related parameters. In other words, the vast majority of customers are more likely to purchase apples with higher "internal comprehensive quality". The advantages and disadvantages of internal comprehensive quality represent the overall status of internal quality at Apple. The question of whether it is feasible to completely and precisely assess the internal quality of an apple has always been one that researchers place a greater emphasis on.

Firmness, crispness, moisture content (MC), sugar content (often reported as soluble solids content), and acidity (typically represented as pH) are the five key factors that define the internal quality of an apple. These primary internal quality characteristics have been successfully detected by nondestructive detection technology in multiple studies over the last ten years [6–11], accurately reflecting the internal quality of an apple. Because of its benefits of simplicity, convenience, high efficiency and speed, and real-time online detection, nondestructive detection technology based on spectroscopy has been widely employed

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in fruit quality testing. Partial least squares regression (PLSR), least squares support vector machine (LSSVM), and BP neural network were used to establish the prediction models for firmness, MC, soluble solids content (SSC), and pH in apples. The characteristic wavelengths were extracted using hyperspectral imaging technology in conjunction with successive projection algorithm (SPA), uninformative variable elimination (UVE), and SPA [10]. Wang et al. [8] explored the quality characteristics of "Fuji" apples in 3 months of late development, including SSC, firmness, pH, and MC. LSSVM and the extreme learning machine (ELM) were used to create the prediction models for these properties utilizing near-infrared diffuse reflectance technology. Ni et al. [11] developed a quantitative prediction model of sharpness. Spectra were used to quickly and nondestructively determine whether an apple was crisp by employing optical fiber spectroscopy. In addition, studies have shown that the rapid nondestructive detection of apple crispness quality could be achieved using spectroscopic technology, and the detection of apple crispness will be more accurate after combining machine vision technology [12]. However, the existing literature all studied the detection of a single internal quality parameter by near infrared spectroscopy, which can only reflect apple's internal quality from a certain, but cannot reflect the internal comprehensive quality of apples, and the research on the internal comprehensive quality of apples has not been reported yet. Therefore, it will be helpful for nondestructive testing of apple quality to build an internal comprehensive quality evaluation and prediction model.

The length of time that apples can be stored depends on their interior overall quality. Apple quality variations during storage have a significant impact on sales quality and customer happiness. Therefore, researchers need to investigate whether the inside quality of an apple can be analyzed thoroughly and properly as well as how this internal quality impacts how apples are stored. Food quality fluctuates throughout time, and kinetic modeling connects these variations to physical, chemical, metabolic, microbiological, and other activities that take place [13]. The quality of fruits, vegetables, seafood, and liquids during storage has been assessed using kinetic modeling [13,14]. Shen et al. employed visible light and near-infrared spectroscopy (Vis/NIR) for online discrimination of the refrigeration duration of strawberry fruits. They also predicted the post-harvest quality of these strawberries. Utilizing partial least squares discriminant analysis, they achieved a classification accuracy ranging from 93.3 % to 97.4 %, providing a foundation for post-harvest storage strategies for strawberries [15]. Pérez-Marín et al. used near-infrared reflectance spectroscopy (NIRS) to categorize the quality of 220 whole nectarines across various postharvest storage durations. With the aid of partial least squares 2-discriminant analysis (PLS2-DA), they correctly classified between 86 % and 96 % of the samples, thereby validating the feasibility of using near-infrared spectroscopy to monitor pre- and post-harvest quality parameters of nectarines [16]. da Silva Ferreira et al. introduced a method for the rapid, non-destructive assessment of dragon fruit quality using low-cost near-infrared spectroscopy (NIRS) and electronic nose (e-nose) equipment. They conducted internal quality evaluations of dragon fruit samples from different periods, confirming that both near-infrared spectroscopy and the electronic nose can effectively discern the internal quality of dragon fruit at various stages [17]. de Moraes et al. utilized a portable near-infrared spectrometer, operating within the 900–1700 nm range, to analyze 177 carambola fruits from two clones and four ripening stages (MS1, MS2, MS3, and MS4). They examined attributes such as color, total solubility, ascorbic acid content, moisture, pH, and titratable acidity. The carambola fruits were classified with an accuracy of 84.2 %. This research underscored the utility of the portable near-infrared spectrometer in classifying carambola based on maturity stages and offers valuable insights for subsequent carambola quality studies [18].

Additionally, kinetic modeling has been used in earlier studies to estimate fruit shelf life, namely apple fruit [19–21]. Based on a zero-order kinetic model, the storage time of fresh jujube stored at room temperature was predicted to be 8 d for yeast infections less than 10 CFU g^{-1} [19]. By developing a quantitative prediction model, a kinetic model for MC, and establishing a correlation between storage time and spectral reflectance before to and after storage, Liu et al. [20] investigated the prediction of MC and the shelf life of apples stored at ambient temperature. Furthermore, Ni et al. [21] used a polynomial kinetic model to find that the optimal freshness of Fuji apple crispness could be preserved for five weeks at ambient temperature and eight weeks under refrigeration. However, to the best of the authors' knowledge, there isn't any research that uses kinetic modeling to examine the internal comprehensive quality of apples, and the prediction of the storage time of apples only takes into account one internal quality component, like crispness and MC.

Therefore, this study will explore the evaluation and prediction of the internal comprehensive quality of apples, and combine with kinetic modeling to study the variations of the internal comprehensive quality of apples during storage and the overall and accurate prediction of storage time. The objectives of this study were: (a) to evaluate the internal comprehensive quality of apples, (b) to quickly and nondestructively predict the internal comprehensive quality of apples, and (c) to predict the storage time of apples.

2. Materials and methods

2.1. Sample preparation

The Luochuan "Fuji" apples selected as the experimental samples were purchased from a local store in Yangling, Shaanxi Province, China, in November 2021. The apples were shipped back to the laboratory on the same day, and then stored in an artificial climate chamber (PGX-250B, Shanghai Kuntian Experimental Instrument Co., Ltd., Shanghai, China) under a temperature of 20 ± 1 ℃ at 35 % relative humidity. Measurements were taken initially and at 5-d intervals during the 50-d storage period. During the detection process, firstly, the spectral information of apples was collected, and then the internal quality parameters of apples were measured by corresponding instruments. When collecting spectral information, three detection points were randomly and evenly selected to collect the spectral information of the apple at the equatorial position of the apple. The internal indexes of the apple were measured at the detection points, and the measuring order was firmness and crispness, SSC, pH and MC. Before each measurement, 10–15 apple fruits (one group) without mechanical damage were washed with tap water to remove foreign matters on the surface and wiped dry. Then, they were labeled and used to collect spectra and measure internal quality. Spectra were acquired from all apple samples and internal quality was measured from one group of samples in each test. Specifically, the spectra of all groups were obtained and the internal quality of the first group was measured in the initial test. In the second test, the spectra of the remaining groups were collected and the internal quality of the second group was measured, and so on. Each sample group containing internal quality data and corresponding spectra data was employed to develop a prediction model. All spectra data and internal quality data of the tenth group of samples were applied to build kinetic models. A total of 132 apple samples were employed in the study.

2.2. Experimental methods

2.2.1. Spectral acquisition

The spectra were acquired using a laboratory-developed optical fiber spectral system in reflectance mode. The system included a computer (Lenovo Legion Y7000 2019 PG0, Lenovo Group, Ltd., Beijing, China) equipped with SpectraSuite (version 2.0.158, American Ocean Photology Company, Florida, USA), an optical fiber spectrometer (USB4000-VIS-NIR, Ocean optics, Florida, USA), a light source, an optical fiber, an optical fiber probe, a conveyor belt, and a stepper motor. Fig. 1 presents the structure of the system.

The optical fiber spectrometer was preheated for 20 min at room

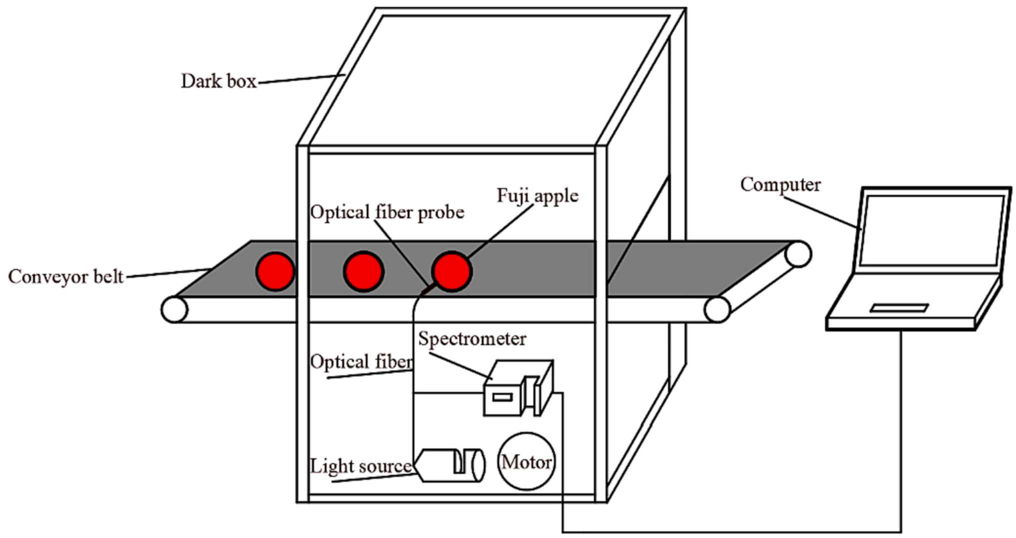

Fig. 1. Structure of the optical fiber spectroscopy system.

temperature (approximately 20 ℃) and calibrated with a standard white panel to eliminate the interference of the light source. The detection wavelength range of the optical fiber spectrometer is 350–1000 nm with a spectral resolution of 1.5–2.3 nm full width at half maxima (FWHM), and the integration time is from 3.8 ms to 10 s with a 0.21 nm sampling interval [11]. When collecting spectra, the parameters of the SpectraSuite software were first set as follows: the integration time, the average number of scans, and smoothness were set as 10 ms, 15 and 10, respectively [11]. Then the optical fiber spectroscopy system was turned on, and the apples to be tested were placed on the conveyor belt. When the equatorial surface of an apple sample was aligned with the optical fiber probe, a spectral curve was collected and recorded. There were three points on the equatorial position of each apple sample where spectra were collected, and the spectral value of the apple was the average of spectra obtained from three points distributed along the apple equator [22,21].

Spectra were acquired from all apple samples and internal quality was measured from one group of samples in each test. Specifically, the spectra of all groups were obtained and the internal quality of the first group was measured in the initial test. In the second test, the spectra of the remaining groups were collected and the internal quality of the second group was measured, and so on. Each sample group containing internal quality data and corresponding spectra data was employed to develop a prediction model. All spectra data and internal quality data of the tenth group of samples were applied to build kinetic models.

2.2.2. Measurement of internal quality parameters

After the spectral acquisition of each apple, a peeler was used to remove the peels [23]. A piece of cylindrical pulp cut through the equator from the core to exocarp (with a diameter and length of 10 mm) was obtained from the spectral collection position of an apple sample, and then the firmness and crispness of the apple were measured with a texture analyzer (TA.XT PLUS/50, Stable Micro Systems, London, UK). The specific parameters of the texture analyzer were set as follows: compression speed of 1 mm/s; the amount of compression of 80 %; and trigger force of 50 N [12]. Regarding firmness and crispness, the maximum force and the first force peak in the TPA texture map were defined as firmness and crispness, respectively [24,25]. After firmness and crispness measurements, the appropriate amount of pulp adjacent to the site for firmness and crispness measurements was put into garlic to extract juice for measuring SSC using a digital refractometer (PAL-1, Atago Co., Ltd., Tokyo, Japan). Then about 2–10 g pulp adjacent to each site for firmness and crispness measurements were used to determine MC determination for 12 h at 70 °C in an electric blast drying oven (WG-71, Tianjin Tester Instrument Co., Ltd., Tianjin, China) until a constant weight of the pulp was reached. Finally, the left pulp of each apple was peeled and placed in the garlic press to squeeze juice, which was obtained in a 10 ml beaker for determining pH with a pH meter (PHSJ-3F, Shanghai Electrical and Instrument Analysis Instrument Co., Ltd., Shanghai, China). The firmness, crispness, MC, SSC and pH of each apple sample at the three spectral collection positions were measured, and an average of three measurements of each parameter was taken as the results of the parameter for the apple.

2.3. Chemometric data processing

2.3.1. Sample partitioning

The joint X-Y distance (SPXY) method is a sample set partitioning algorithm extended from the classic Kennard-Stone (KS) algorithm by Galvão et al [26]. Unlike the KS algorithm, which only calculates the inter-sample Euclidean distances between x and vectors, the algorithm encompasses both x- and y-variables in the calculation of distances between samples. Some work has shown that SPXY can improve the predictive performance of models [27]. In the study, SPXY was introduced to partition sample sets according to the ratio of 3:1, indicating 99 samples in the calibration set and 33 samples in the prediction set. The calibration set was applied to develop stable and reliable models, while the prediction set was used to validate the model performance.

2.3.2. Spectral preprocessing

The purpose of spectral preprocessing is to filter out the noise information in spectra and eliminate or reduce the scattering effects caused by multicollinearity and baseline offset, inhibiting the complexity of models and improving their robustness [28]. At present, spectral preprocessing methods such as smoothing, derivative, multiplicative scatter correction (MSC) and standard normal variate (SNV) are widely used [29]. In the study, smoothing, first derivative (first D), standard normal variate (SNV) and their combinations were used to preprocess raw spectra and establish the models. The pretreatment methods that were optimal for each internal quality parameter and the internal comprehensive quality index were selected for the subsequent models.

2.3.3. Variable selection strategy

Competitive adaptive reweighted sampling (CARS) is a new variable selection strategy based on Monte Carlo sampling and PLS regression coefficients proposed by Li et al [30]. For CARS, effective wavelengths are defined as the wavelengths with large absolute coefficients in a multivariate linear regression model, such as PLS. During each sampling run, CARS performs four successive procedures including Monte Carlo model sampling, exponentially decreasing function (EDF)-based enforced wavelength selection, competitive wavelength selection by adaptive reweighted sampling (ARS), and root-mean-square error of cross-validation (RMSECV) computation for each subset [31]. Some subsets of variables are chosen by the CARS algorithm through many sampling runs and then the subset with the smallest RMSECV value is selected as the optimal wavelength combination. The CARS method was carried out in the MATLAB software (version R2019b, MathWorks, Natick, MA).

2.3.4. Partial least squares regression

Partial least squares regression (PLSR) is a bilinear modeling method that combines the advantages of principal component analysis (PCA) and multiple linear regression (MLR) and has been widely employed in near-infrared spectroscopy analyzes. By using PLSR, the original independent information in X (spectral data matrix) is projected into a small number of latent variables (LVs) to simplify the relationship between X and Y (target quality property matrix) [32,33], making it easier to predict. When applying PLSR for modeling, it is particularly important to determine the optimal number of LVs participating in the regression models. If fewer LVs are selected, models will be insufficiently fitted; if more LVs are chosen, models will be over fitted [12]. Based on the minimum value of the predicted residual error sum of squares (PRESS), the optimal LVs of the PLSR models are determined via the leave-one-out cross-validation method. The PLSR modeling was employed using Unscrambler X software (version 10.4, Camo Process AS, Oslo, Norway) in this study.

2.3.5. Model assessment

Several parameters were used to evaluate the model performance based on statistical tests in this study. These parameters included the determination coefficients of calibration (R_c^2) and prediction (R_p^2), root mean square errors of calibration (RMSEC) and prediction (RMSEP) as well as residual predictive deviation (RPD). The RPD, which is the ratio of the standard deviation (SD) for the prediction set to the RMSEP, was applied to assess the prediction abilities of models. A good model should have high values of R^2 and RPD, low values of RMSE's, as well as a small difference between RMSEC and RMSEP [34,35].

According to Mireei et al [36], three quality categories were defined to explain the reliability and performance of models:

(1) a very reliable and very good performance of the model, in which the RPD ranges from 6.5 to 8.0;
(2) a reliable and good performance of the model, in which the RPD ranges from 5.0 to 6.4;
(3) a non-reliable and poor performance of the model, in which the RPD is less than 5.0.

2.4. Reference data analysis

2.4.1. Correlation analysis between different quality parameters

To evaluate the property and extent of relationships that exist among internal quality parameters of apples, Pearson's correlations between firmness, crispness, MC, SSC and pH were calculated, and the significance of their associations was tested with a *t*-test at a significance level of 0.05 and 0.01 [37]. Before correlation analysis using SPSS software (version 24, IBM, Armonk, USA), internal quality parameters data were positive and standardized to maintain the consistency of data properties and eliminate the dimensional influence of data, respectively.

2.4.2. Determination of the weight of each quality parameter

Analytic Hierarchy Process (AHP) is a multiple-attribute decision-making method, which is based on three principles, i.e. hierarchical structure, the pairwise comparative judgment of elements at the same level and the synthesis of priorities. The method can address the question of how to determine the relative importance of a set of factors in multi-criteria problems, thereby combining difficult-to-quantify qualitative judgments with quantitative measures [38,39]. In this study, AHP was applied to determine the weight of each internal quality parameter to construct an internal comprehensive quality attribute. The used AHP was completed on MATLAB software (version R2019b, MathWorks, Natick, MA). A pairwise comparison matrix was constructed by a nine-point scale method to calculate the weight coefficients, and the consistency was tested with the consistency index (CI) and the consistency ratio (CR) calculated according to Eqs. (1)-(2) respectively, as follows:

$$CI = \frac{\lambda_{max} - n}{n - 1} \tag{1}$$

$$CR = \frac{CI}{RI} \tag{2}$$

where λ_{max} and n represent the maximum eigenvalue and order of the pairwise comparison matrix, respectively, and RI represents the average random consistency index corresponding to n. When $CR < 0.10$, the pairwise comparison matrix can be considered acceptable; otherwise, the pairwise comparison matrix needs to be adjusted and modified.

2.5. Chemical kinetic analysis

The quality of foods changes over time, so quality parameters related to food quality governed by chemical, biochemical, microbial and physical changes are not constant. It is considered that modeling related quality changes to chemical, biochemical, microbial and physical processes that occur in food is kinetic modeling [13]. Kinetic modeling indicates that these quality changes can be captured from mathematical models including kinetic characteristics, such as activation energy, reaction rate constants and so on. Zero-order and first-order reactions are frequently reported for quality variations in most high-quality foods. Second-order and higher-order reactions are sometimes reported in some food changes [13,40]. The kinetic reaction equations are calculated as follows:

$$zero-orderreaction:\ C = C0 - kt \tag{3}$$

$$first-orderreaction:\ C = C0 \times e^{-kt} \tag{4}$$

$$higher-orderreaction:\ C = k0t^n + k1t^{n-1} + \ldots + kn - 1t + C0 \tag{5}$$

where C is the quality indicator value; C_0 is the initial quality indicator value; t is storage time; and $k_0 \ldots k_{n-1}$ are the reaction rate constants.

3. Results

3.1. Statistical analysis of quality parameters

3.1.1. Description of measurement data

The statistics of firmness, crispness, MC, SSC and pH values of used 132 apple samples in the calibration and prediction sets are presented in Table 1. Statistics for these quality parameters included the number of samples, range, mean value and standard deviation. We can see from Table 1 that the range of the calibration set covers that of the prediction set for each quality parameter, which can help to develop prediction models with strong stability and robustness. In addition, the standard deviation of each quality parameter was 18.84–23.68 % of the difference between the maximum and minimum values of that parameter in the calibration set, and 20.20–27.45 % of the difference between the maximum and minimum values of that parameter in the prediction set. The standard deviations of the calibration and prediction sets are not significantly different, so the distribution of the samples is appropriate

Table 1
Statistics of apple's internal quality parameters in the calibration and prediction sets.

Subset	Number of samples	Quality parameter	Minimum	Maximum	Mean	Standard deviation
Calibration set	99	Firmness ($kg \cdot cm^{-2}$)	2.69	5.98	4.17	0.67
		Crispness ($kg \cdot cm^{-2}$)	2.20	5.65	3.73	0.65
		MC (%)	81.08	87.49	84.58	1.44
		SSC (°Brix)	11.70	17.60	13.96	1.24
		pH	3.82	4.58	4.15	0.18
Prediction set	33	Firmness ($kg \cdot cm^{-2}$)	3.48	5.38	4.22	0.43
		Crispness ($kg \cdot cm^{-2}$)	3.11	5.14	3.79	0.41
		MC (%)	82.89	86.57	84.64	1.01
		SSC (°Brix)	12.60	15.80	14.05	0.84
		pH	3.83	4.34	4.08	0.14

for either the calibration or prediction sets.

3.1.2. Pearson correlation analysis between quality parameters

The results of correlation and significance analysis of apple's internal quality parameters are shown in Table 2. There is no significant correlation among most of the quality properties, and extremely significant or significant correlations exist only between a few pairwise quality traits. Crispness and SSC were positively correlated with firmness ($^{**}P < 0.01$; $^{*}P < 0.05$), with correlation coefficients of 0.233 and 0.199, respectively. There were significant negative relationships between MC and firmness, and between SSC and MC ($^{**}P < 0.01$; $^{*}P < 0.05$), and the correlation coefficients were −0.431 and −0.153, respectively. Although there are very significant or significant correlations between these quality indicators, the absolute values of their correlation coefficients were far less than 0.7, and the correlations were weak. Therefore, it can be considered that the five internal quality indexes are independent of each other, and the linear combination of these internal quality parameters is used to construct an internal comprehensive quality attribute that represents the internal quality information of apples.

3.2. Characterization of internal comprehensive quality

3.2.1. Determination of weight for single quality parameter using AHP

There were a total of 5 experts (all professors of agricultural engineering or horticulture) who participated in the comparisons of the importance of quality traits in the study. According to their negotiated scoring results, the pairwise comparison matrix and weight calculation results from AHP are described in Table 3. As can be seen from Table 3 that the CR value (0.0461) was less than 0.1, that is, the pairwise comparison matrix passes the consistency test and is thought to have good consistency, so the weight coefficients calculated by the constructed pairwise comparison matrix is reliable. The weight calculation results indicate that SSC is considered to be the most important quality index among the five main internal quality indicators, which had the highest weight of 0.4553. Firmness and pH had the lowest weights, both of 0.0704, indicating that they are relatively less important properties among the main internal quality parameters of apples.

Table 3
Pairwise comparison matrix and weights from AHP for the evaluation hierarchy.

	Pairwise comparison matrix					Weight
	Firmness	Crispness	MC	SSC	pH	
Firmness	1	1/3	1/5	1	1/3	0.0704
Crispness	3	1	1/3	3	1/3	0.1561
MC	5	3	1	5	3	0.2478
SSC	1	1/3	1/5	1	1/3	0.4553
pH	3	3	1/3	3	1	0.0704
		$\lambda_{max} = 5.2067, CI = 0.0517, RI = 1.1200, CR = 0.0461$				

3.2.2. Construction of an internal comprehensive quality attribute

The results of Pearson correlation analysis and the weights determined by AHP for single quality index demonstrated that the internal quality parameters such as firmness, crispness, MC, SSC and pH are approximately orthogonal to each other, and the determined weights (also for the degree of contribution to the construction of an internal comprehensive quality attribute) were 0.0704, 0.1561, 0.2478, 0.4553 and 0.0704, respectively. The expression of the constructed internal comprehensive quality indicator of apples is as follows:

$$ICQ = 0.0704F + 0.1561C + 0.2478MC + 0.4553SSC + 0.0704pH \tag{6}$$

where ICQ represents the constructed internal comprehensive quality trait, F ($kg \cdot cm^{-2}$), C ($kg \cdot cm^{-2}$), MC (%), SSC (°Brix) and pH represent the firmness value, crispness value, MC value, SSC value and pH value after positive normalization and standardization, respectively.

3.3. Prediction of internal comprehensive quality

3.3.1. Development of a calibration model

The collected spectra in the entire wavelength range of 600–900 nm were selected variables using the CARS algorithm with the following parameter settings: spectral matrix size of 132 × 1588; internal comprehensive quality index matrix size of 132 × 1; maximal extraction

Table 2
Correlation and significance analysis between internal quality parameters of apples.

		Firmness	Crispness	MC	SSC	pH
Correlation	Firmness	1.000				
	Crispness	0.233**	1.000			
	MC	−0.431**	−0.132	1.000		
	SSC	0.199*	0.083	−0.153*	1.000	
	pH	−0.063	0.126	−0.080	0.065	1.000
Significance (one-tailed)	Firmness					
	Crispness	0.005**				
	MC	0.000**	0.074			
	SSC	0.014*	0.181	0.046*		
	pH	0.244	0.084	0.192	0.239	

principle of 30; cross-validation group number of 2; center pretreatment method; and Monte Carlo sampling runs number of 50. The optimal LVs were selected to achieve the global minimum of the RMSECV curves using the original version of CARS. Fig. 2 depicts the process of selecting variables for the internal comprehensive quality attribute using CARS. As the number of sampling runs increased, the variables with large contribution rate to models were retained while those with small contribution rate to models were eliminated, and the number of variables gradually decreased. In the meantime, the RMSECV of models gradually decreased as the number of sampling runs increased to 26, where it reached a minimum (0.0015). The variable subset in this sampling was selected as the optimal spectral variable combination for the internal comprehensive quality properties, and it contained 53 variables (effective wavelengths). Fig. 3 displays the result of selecting effective wavelengths for internal comprehensive quality traits based on the CARS algorithm.

Near infrared spectroscopy is sensitive to the concentration of organic materials, which involves the response of molecular bonds of C–H, O–H and N–H. As can be seen from Fig. 3, the low values of reflectance in the range of 600–610 nm, i.e., the high absorbance, might be attributed to the O–H second overtone from carbohydrates and water. The values of reflectance decreased rapidly at the wavelength of 800–870 nm, which was attributed to C–H stretching second overtone from carbohydrates (fructose, sucrose, and glucose).

The effective wavelengths selected by CARS were used as the input of modeling, while the constructed internal comprehensive quality attribute values using AHP were the output of modeling, and a PLSR prediction model (CARS-PLSR) was established. As shown in Fig. 4, as the number of LVs in the CARS-PLSR model increased, PRESS quickly decreased to a constant value, while the explained X total variance and explained Y total variance gradually accumulated. The determined optimal number of LVs was 7 When PRESS reduced to a minimum value of 0.27, and the model explained 85.80 % spectral variance, while the top 7 LVs compressed 96.37 % property variance.

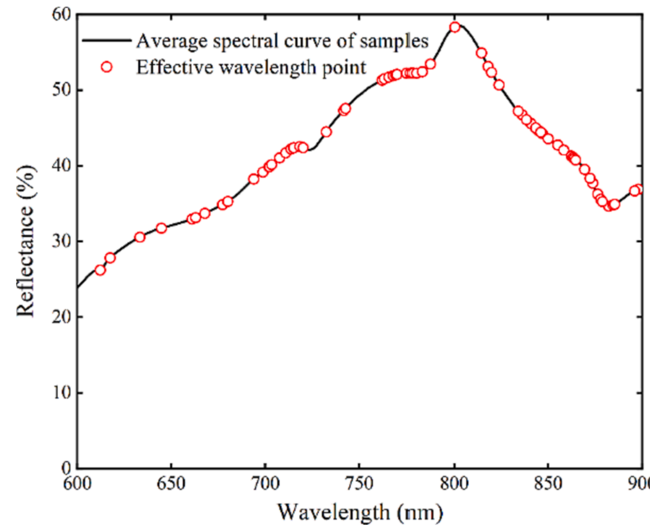

Fig. 3. The effective wavelength selection result of CARS for the internal comprehensive quality indicator.

Fig. 5 suggests the measured values of the internal comprehensive quality attribute against predicted ones for calibration and prediction sets of the PLSR model developed with effective wavelengths. The R_c^2 and R_p^2 of the established CARS-PLSR model were 0.9804 and 0.9419, respectively, with an RMSEC and RMSEP of 0.0020 and 0.0023, respectively, and an RPD of 5.77. The CARS-PLSR model had high values of R^2, low values of RMSE's, a smaller difference between RMSEC and RMSEP, as well as the RPD value greater than 5.00, indicating that the established prediction model has an excellent performance and can greatly evaluate the internal comprehensive quality indicators of apples. Therefore, the equation of the established CARS-PLSR model is as follows:

$$\begin{aligned} I = & -0.2679X612.27 + 0.4219X617.49 - 0.2656X633.30 + \ldots - 0.5149X873.64 \\ & -0.5562X882.17 - 0.0981X898.05 + 6.3825 \end{aligned} \tag{7}$$

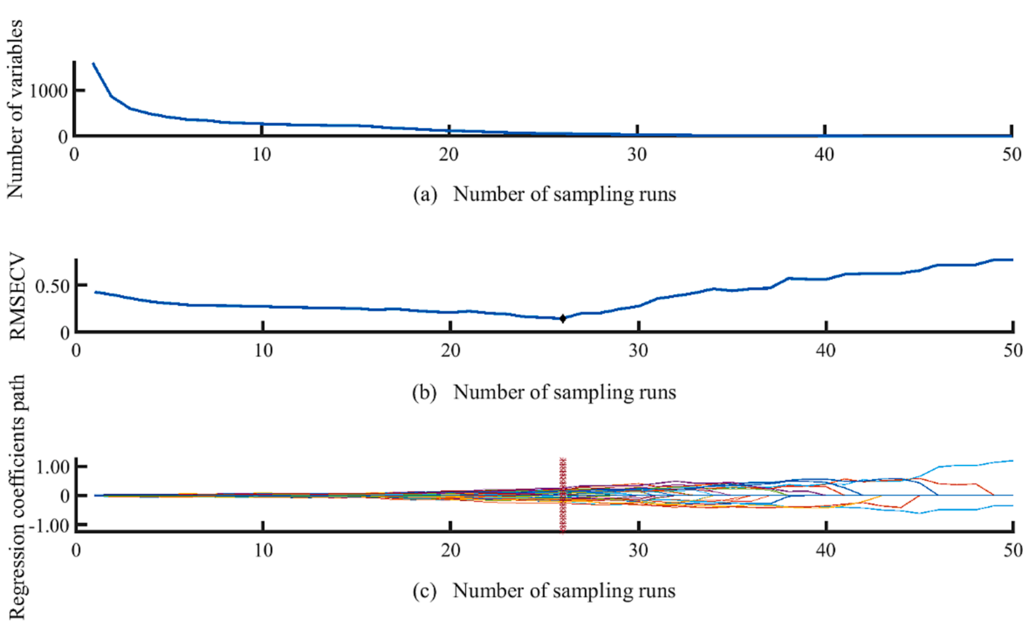

Fig. 2. Spectral variable selection by competitive adaptive reweighted sampling (CARS) algorithm.

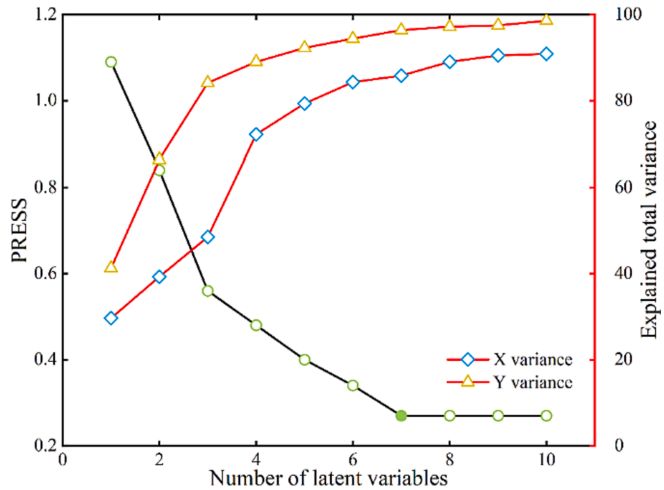

Fig. 4. Changed PRESS and explained the total variance in the number of LVs in the CARS-PLSR model. The Green solid circle represents the point at which the optimal number of variables was determined. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

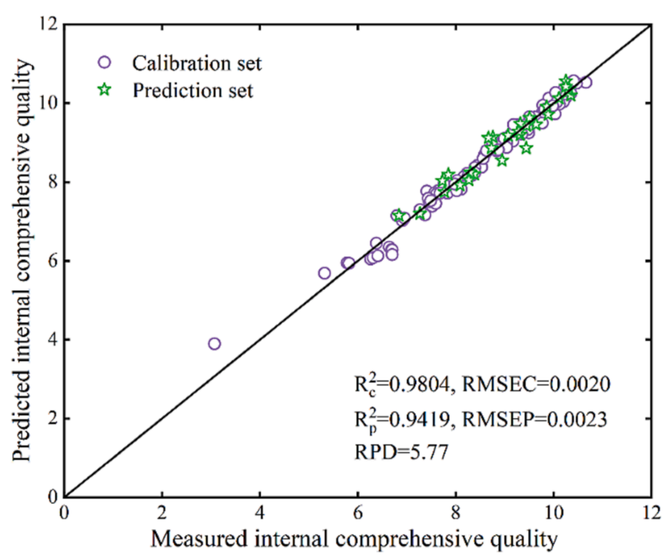

Fig. 5. The measured values of the internal comprehensive quality attribute against predicted ones for calibration and prediction sets in the CARS-PLSR model.

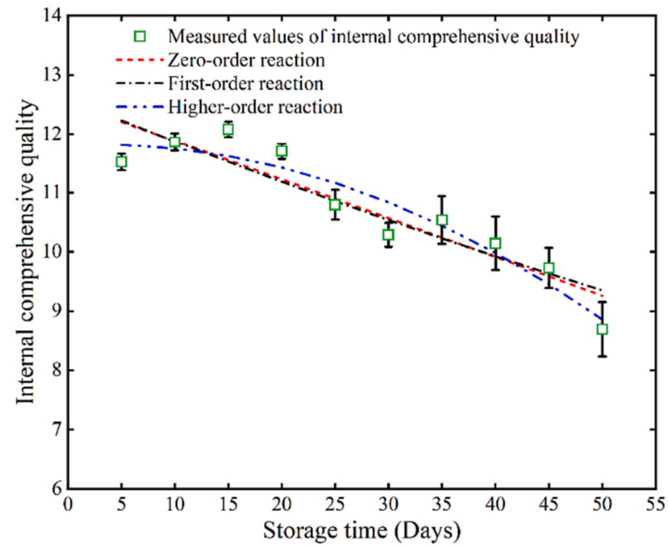

Fig. 6. Results of fitting the internal comprehensive quality over storage time using zero-, first- and high-order reactions.

where I represent the internal comprehensive quality attribute; and $X612.27 \ldots X898.05$ represent spectral reflectance at different effective wavelengths, respectively.

3.3.2. Establishment of kinetic models

The characteristics of the experiment suggested that the internal quality values were destructively measured by just one group of samples in each test to determine the internal comprehensive quality. The sample group in the last test (the tenth group) participated in the whole experiment, and their spectra were collected in every test. For the tenth group of samples, the internal comprehensive quality values predicted by the established CARS-PLSR model were used as their measured values. In addition, according to the variations in internal comprehensive quality under different storage times, the relationship (which describes the variations in internal comprehensive quality with the storage time) were explored by comparing three kinetic modes (zero-, first-, and higher-order reactions). Fig. 6 shows the results of fitting the internal comprehensive quality over storage time using three kinetic reactions. The internal comprehensive quality first increased and then decreased with the prolongation of storage time. All three kinetic reactions could well describe the variations in internal comprehensive quality with storage time, and the higher-order reaction was more in line with this trend. Therefore, it was reasonable to believe that the variations in the internal comprehensive quality of apples with storage time followed higher-order reaction.

Kinetic models were built based on the three kinetic reactions, and Table 4 compares the prediction results of these kinetic models with the internal comprehensive quality. The three kinetic models all exhibited higher correlation coefficient (R) and lower RMSE, which can well predict the internal comprehensive quality. Compared with the zero- and first-order kinetic models, the higher-order kinetic model proved a higher R and lower RMSE, and its prediction performance had obvious advantages. Therefore, the high-order kinetic model could accurately evaluate the internal comprehensive quality of apples, and the expression of the model is:

$$\begin{aligned} I_t = & -0.0019t^2 - 0.0027t - 0.2679X612.27t0 + 0.4219X617.49t0 - 0.2656X633.30t0 + \ldots - 0.5149X873.64t0 \\ & -0.5562X882.17t0 - 0.0981X898.05t0 + 5.7792 \end{aligned} \quad (8)$$

where I_t represents the internal comprehensive quality for t d; t represents storage time (d); and $X612.27t0 \ldots X898.05t0$ represent the spectral reflectance at different effective wavelengths before storage, respectively.

Table 4
Prediction results on the internal comprehensive quality using three different kinetic models.

Items	Correlations	R	RMSE	Kinetic models
Zero-order reaction reaction reaction	Linear	0.9281	0.0052	$I_t = 1.4404I_0 - 4.8998 - 0.0902t$
First-order reaction	Exponential	0.9137	0.0057	$I_t = (I_0 - 2.0029)e^{-0.0082t}$
Higher-order reaction	Parabolic	0.9620	0.0038	$I_t = -0.0019t^2 - 0.0027t + I_0 - 0.6033$

3.4. Prediction of storage time

The internal comprehensive quality variations of fresh apples could be directly reflected by the length of storage time, that is, the two followed a higher-order kinetic reaction. Therefore, storage time could be determined according to the variations in internal comprehensive quality pre- and post-storage. Specifically, both the prediction model determined by Eq. (7) and the kinetic model proposed by Eq. (8) could be employed to evaluate the internal comprehensive quality (storage for t d), and we developed Eq. (9) to predict the storage time of apples by combining Eqs. (7) and (8) as follows:

$$t = 22.9416\sqrt{\begin{pmatrix} -0.2679(X612.27t0 - X612.27t) + 0.4219(X617.49t0 - X617.49t) - \\ 0.2656(X633.30t0 - X633.30t) + \ldots - 0.5149(X873.64t0 - X873.64t) - \\ 0.5562(X882.17t0 - X882.17t) - 0.0981(X898.05t0 - X898.05t) - 0.6033 \end{pmatrix}} - 0.7105 \tag{9}$$

where t represents storage time (d);$X612.27t0 \ldots X898.05t0$ represent the spectral reflectance at different effective wavelengths pre-storage, respectively; and $X612.27t \ldots X898.05t$ represent the spectral reflectance at different effective wavelengths following storage for t d, respectively.

As seen from Equation (9), storage time could be finally determined by the difference in the spectral reflectance of apples pre- and post-storage. Fig. 7 reveals the prediction results of storage time for the tenth group (12 samples in total) using Eq. (9). The R_c^2 and R_p^2 of the prediction model were 0.9059 and 0.8957, respectively, as well as the RMSEC and RMSEP less than 5 d. The prediction accuracy of the model was good, and it is considered that the prediction model can well meet the actual needs of accurately predicting the storage time of apples within the acceptable error range. It is worth noting that since the time interval of 5 d was used as a test period in the study, the acceptable error range of the predicted storage time was considered that it should not exceed 5 d before and after the actual measured storage time.

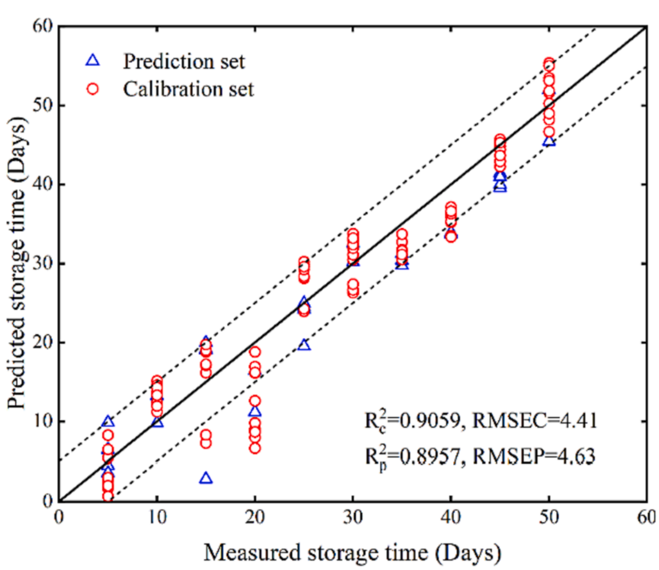

Fig. 7. The measured values of storage time against predicted ones for calibration and prediction sets using the proposed model.

4. Discussion

In order to characterize the comprehensive internal quality of apples to meet consumer needs, this study constructed an internal comprehensive quality index by selecting the five main quality parameters of firmness, crispness, MC, SSC and pH, which are the most significant for internal quality. Compared with a single internal quality parameter, this index shows the internal quality of apples more comprehensively, and can reflect the texture and taste of apples in a more diverse manner, which is undoubtedly novel and exciting for expressing the comprehensive internal quality of apples. However, since the evaluation of apple quality varies from person to person, there is currently no unified evaluation criterion. Therefore, although the internal comprehensive quality index constructed in the study can comprehensively evaluate the internal quality of apples, there is still some subjectivity in constructing this index. This subjectivity mainly focuses on the fact that the weights of the main internal quality parameters to the construction of the internal comprehensive quality index need to be determined artificially. Due to the wide variation in individual taste preferences, the weights assigned by consumers and researchers from various groups may be highly varied, which could cause inconsistency or even be at odds with the work we do. But we don't think this will affect the innovation and contribution of this study: we mainly prove that it is feasible to comprehensively evaluate the internal quality of apples; according to the work of this study, various internal comprehensive quality indexes can be constructed by adjusting these weights to meet the specific quality preferences and needs of different consumer groups and researchers.

To predict the interior comprehensive quality of apples, we created quantitative and kinetic models in this work. The calibration model was able to accurately estimate the internal comprehensive quality of apples based on the spectral data. Then, using the predicted internal comprehensive quality, kinetic models for apples under various conditions were developed. The kinetic models are also proved to accurately predict the internal comprehensive quality of apples under the premise of the known initial spectral information and storage time of apples. Both the two models can predict the internal comprehensive quality of apples. The difference is that the calibration model relies on spectral characteristics to predict internal comprehensive quality online in real time, and kinetic models can not only describe specific variations in internal comprehensive quality during storage period but also predict future internal comprehensive quality values of samples with the help of time series characteristics [20,21]. By comparing the calibration model with kinetic models, the internal comprehensive quality of the same apples was precisely evaluated, and the results were within the acceptable deviations [20,21]. Furthermore, the dependency of kinetic models on time series allows them to predict the storage time of apples.

The storage time of apples depends on variations in the apple's internal quality, and previous studies have failed to identify a potential relationship between the two. This study clarified that variations in apple's internal quality and the storage time of apples followed a higher-order kinetic reaction, and determined a prediction model by combining the calibration model and kinetic model established for internal comprehensive quality. The storage time of apples can be determined by the prediction model only using the difference in the spectral reflectance of apples pre- and post-storage. Since our work fully takes into account the synergistic effect of variations in the apple's internal quality attributes during the storage process on the storage time, the prediction of the storage time of apples is very accurate and can better meet the actual needs. However, there is still a certain limitation in our work, which is mainly reflected in exploring the influence of the comprehensive changes of apple's internal quality on the storage time of apples. Only variations in the physicochemical quality indicators of fruits are analyzed, and variations in the physiological quality attributes of fruits are not considered. Therefore, there is still a certain deviation between the predicted and the actual storage time of apples. Employing a comprehensive analysis combined with the detailed variations in physiological indicators of apple fruit during storage, which are of great significance for the promotion of food quality research, is reserved for future work on food quality storage.

5. Conclusions

In this study, an internal comprehensive quality index was constructed to characterize the comprehensive internal quality of apples, and a calibration model and a high-order kinetic model were developed to evaluate the internal comprehensive quality of apples. In addition, a prediction model was established to predict the storage time of apples.

(1) An internal comprehensive quality index was constructed. An internal comprehensive quality index was constructed by five internal quality parameters: firmness, crispness, MC, SSC and pH. The weights of firmness, crispness, MC, SSC and pH in the constructed internal comprehensive quality index were 0.1561, 0.0704, 0.2478, 0.4553 and 0.0704 respectively. The constructed internal comprehensive quality index realizes the comprehensive and quantitative characterization of apple's internal quality.
(2) The calibration model and high-order kinetic model of the internal comprehensive quality index were established. According to the spectral data of apples, internal comprehensive quality can be evaluated using the calibration model, and the evaluated internal comprehensive quality can be used to develop kinetic models under different conditions. On the premise of knowing the spectral data and storage time of apples during initial storage, internal comprehensive quality can be accurately evaluated by the high-order kinetic model. Both the quantitative model and the high-order kinetic model can evaluate the internal comprehensive quality of apples.
(3) The prediction model of the storage time of apples was determined. The storage time of the tenth group of samples was predicted by the built prediction model. The R_c and R_p of the model were 0.9059 and 0.8957 respectively, and the RMSE was less than 5 d. Within the acceptable error range, it is considered that the prediction model can meet the actual demands of accurately predicting the storage time of apples.

6. Practical application

This study provides a method for determining an apple's internal comprehensive quality and estimating its storage time. According to the proposed method, businesses may easily forecast the storage period of apples to identify whether or not they are safe, in addition to swiftly and accurately evaluating internal comprehensive quality to assess the texture and flavor of apples.

Ethical statements

Ethical Review: This study does not involve any human or animal testing. This study was approved by the Institutional Review Board of Northwest A&F University and conforms to the Declaration of Helsinki, US.

Informed Consent: Written informed consent was obtained from all study participants.

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

Data will be made available on request.

Acknowledgments

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