

Review



Advancements and Applications of Raman Spectroscopy in Rapid Quality and Safety Detection of Fruits and Vegetables

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Abstract: With the improvement in living standards, consumers have become more aware of healthy diets and pay more attention to the quality and safety of fruits and vegetables. Therefore, it is essential to strengthen the research on rapid detection of the quality and safety of fruits and vegetables. This study mainly outlines five Raman spectroscopy techniques. It introduces their principles and advantages and the current research progress of their application in fruit and vegetable quality and safety detection. Based on the characteristic Raman spectroscopy analysis of different fruits and vegetables, researchers found that Raman spectroscopy techniques can quickly and accurately detect classification identification, ripeness, freshness, disease infestation, and surface pesticide residues of fruits and vegetables. In addition, Raman spectroscopy techniques can also detect the content and distribution of material components of fruits and vegetables. This paper also discusses Raman spectroscopy's current technology and application difficulties in fruit and vegetable quality and safety testing. It looks forward to its future development trend, expecting to promote the broad application of Raman spectroscopy in fruit and vegetable quality and safety testing.

Keywords: Raman spectroscopy; fruit and vegetable quality; safety testing; surface pesticide residues; nondestructive testing

1. Introduction

Fruits and vegetables are rich in vitamins and nutrients, and they play a crucial role in maintaining the daily nutritional intake of the human body [1]. However, fruits and vegetables are fresh foods. They are easily affected by external or internal factors during planting, harvesting, transportation, packaging, and storage, resulting in varying degrees of deterioration and disease in their quality. According to the report released by Chinese official institutions, the post-production loss rate of fruits and vegetables in China is 15–20% and 20–25%, respectively, much higher than the developed countries average level. Furthermore, approximately 200 million tons of fruits and vegetables are lost annually during transportation. The spoilage and decay of these products could provide essential nutritional requirements for close to 200 million individuals but instead result in economic losses of 75 billion yuan [2,3]. In addition, the overuse of pesticides by producers to enhance crop production results in residual pesticides in fruits and vegetables, which pose a significant risk to human health, potentially leading to poisoning and various diseases [4-6]. Therefore, developing a rapid, highly accurate, non-destructive fruit and vegetable quality and safety testing technology is highly significant from a practical standpoint. The conventional techniques of assessing the quality of fruits and vegetables mainly rely on sensory evaluation and chemical analysis. However, these methods are vulnerable to interference, exhibit low accuracy, slow detection speed, and require high resource consumption. Therefore, researchers worldwide have continuously explored and developed intelligent non-destructive detection technologies, including Raman spectroscopy,



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Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). near-infrared spectroscopy, RGB vision detection, electronic nose, acoustic characteristics, dielectric properties, and other emerging nondestructive testing technology [7].

Nondestructive testing technologies commonly used today have their advantages. Near-infrared spectroscopy inspection technology analyzes different spectral features of fruits and vegetables within the near-infrared spectrum. Due to its strong penetrating power, it is primarily employed for evaluating the internal quality of fruits and vegetables. RGB vision detection technology analyzes fruits and vegetables based on their image information, offering high efficiency, automation, and other advantages. It is primarily utilized for the inspection of fruit and vegetable appearance. Electronic nose technology, which simulates the olfactory system of animals, can analyze the odor emitted by fruits and vegetables to distinguish their varieties and quality, with the advantages of rapid detection, high precision, non-destructive, etc., mainly for testing the freshness, maturity, and varieties of fruits and vegetables. Acoustic characteristics technology can be utilized to assess the ripeness, freshness, and overall quality of fruits and vegetables by analyzing their acoustic signals. Dielectric property technology is a non-destructive, fast, and efficient method that measures the dielectric properties of fruits and vegetables to assess their water content, hardness, and other important information.

Raman spectroscopy is a detection technique that utilizes the frequency shift and intensity changes of scattered light when a sample interacts with a laser light source, which allows for the acquisition of molecular vibrational information and enables the analysis and identification of the chemical composition, structure, and properties of the sample. Apart from the common advantages of high efficiency and non-destructive properties, Raman spectroscopy is not interfered with by water. It can be detected through the aqueous solution. Raman spectroscopy detects various types and indexes of fruits and vegetables and is suitable for internal and external quality inspection. Due to its high sensitivity and molecular specificity, this technique excels in detecting trace components often missed by other non-destructive testing methods. By analyzing the characteristic vibration frequency of molecules, it can quickly evaluate the quality and nutritional composition of fruits and vegetables while detecting material composition and structure [8]. Furthermore, this technology can detect hazardous substances and surface pesticide residues in fruits and vegetables. Given its potential for development and application, it is a promising solution for assessing fruit and vegetable quality and safety.

This study elaborates on five types of Raman spectroscopy technologies. It summarizes the current research on applying Raman technology for detecting fruit and vegetable quality and safety, mainly including variety classification, qualitative and quantitative quality analysis, spoilage bacteria leaching, drug residue detection, etc. Furthermore, this study discusses the current difficulties in technology, and practical application of Raman technology, and the development trend of its outlook for the future.

2. Overview of Raman Spectroscopy

Raman spectroscopy is a type of molecular scattering spectroscopy discovered by the Indian scientist C.V. Raman in 1928 [9]. The experimental study demonstrated that molecules of matter irradiated by a laser light source exhibit an inelastic scattering effect. This leads to the emission of scattered light spectra with different frequencies from the incident light frequency. When photons from an excitation light source interact with a molecule that acts as the scattering center, most photons undergo Rayleigh scattering, whereby they only change direction and scatter. In contrast, the frequency of light remains the same as that of the excitation light source. In addition, a small number of photons cannot only alter the direction of light propagation and modify the frequency of the light wave, known as Raman scattering. In a given sample, the shift observed in the Raman spectrum remains constant irrespective of the frequency of the incident light. It is determined by the molecular vibrational and rotational energy levels of the substance. As a result of having unique vibrational and rotational energy levels, different substances exhibit distinct Raman shifts, indicating specific vibrational frequencies of functional groups or chemical bonds of the measured molecules, as reflected by the peak positions in Raman spectroscopy. Due to its unique characteristics, Raman spectroscopy can be a powerful tool for structure identification and molecular interaction analysis. Specifically, important information regarding Raman peaks can be extracted to identify distinct structural features and groups, such as C–C, S–S, N–N bonds, etc. [10].

Raman spectroscopy is based on the Raman scattering effect, which generates a unique Raman fingerprint spectrum for each substance molecule. Each peak corresponds to a unique molecular bond vibration, which can facilitate the quantitative and qualitative analysis of target samples at the molecular level by evaluating several parameters, including Raman peak intensity, linearity, number of spectral lines, and peak position [11]. In addition, the Raman scattering of water is very weak and can be used to study samples in aqueous solvents or contained in standard glass vials. Raman spectroscopy's light can be detected through water or glass without significant interference. This versatile technique can also analyze organic and inorganic substances over 50–4000 wavelengths.

3. Types of Raman Spectroscopy Techniques

3.1. Confocal Micro Raman Spectroscopy

Confocal micro-Raman spectroscopy is a technical application that combines the advantages of both Raman spectroscopy and microanalysis techniques. The excitation light focuses the spot into the micron scale space, the sample is analyzed for Raman signal, and the image is reconstructed into a three-dimensional image using a confocal pinhole combined with automatic computerized continuous slice layer scanning. As a result, confocal microscopy outperforms conventional microscopes by suppressing image blurring, offering more precise imaging, achieving high spatial resolution, and enabling more accurate localization and quantitative analysis [12–14].

3.2. Fourier Transform Raman Spectroscopy

The principle of Fourier transform Raman spectroscopy is to collect Raman spectra by using a 1064 nm excitation light source to irradiate the sample, overcoming the interference of fluorescence, and performing Fourier transform technique to collect the signal. Fourier transforms Raman spectroscopy combines Fourier transform technology with near-infrared excitation Raman technology, making it suitable for samples with intense fluorescence, deep color, and instability to light, with the advantages of reducing fluorescence interference, fast scan spectrum, spectral presentation complete, etc. In addition, it has the advantages of reduced fluorescence interference, fast scanning spectra, and complete spectral presentation [15]. In 1986, Hirschfeld and Chase demonstrated practical applications of this technique [16].

3.3. Surface-Enhanced Raman Spectroscopy

Surface-enhanced Raman spectroscopy (SERS) exploits the enhancement of electromagnetic fields between the sample and a rough metal surface upon adsorption or proximity. This phenomenon is known as surface-enhanced Raman scattering and increases Raman signal intensity by a factor of 10^4 – 10^6 , effectively solving the issue of weak Raman signals [17,18]. SERS commonly utilizes noble metals (e.g., Au and Ag) with nanoscale surface morphology as substrates and is widely used for detecting trace analytes due to its high detection sensitivity [19].

3.4. Resonance Raman Spectroscopy

The resonance Raman scattering effect significantly increases the intensity of the Raman band spectral signal almost a million times. The principle is that when the excitation light frequency is close to or overlaps with the electron absorption peak frequency of the target molecule, a resonant state is achieved where the excitation light couples with the molecule's electron, generating the resonance Raman effect [20,21].

3.5. Spatial Offset Raman Spectroscopy (SORS)

The principle of spatially offset Raman spectroscopy is that after the light source excites the tested sample, the Raman signal collection point deviates from the incident focal point by a certain displacement distance to collect the Raman signal. The larger the moving position, the deeper the penetration of the substance to be examined. The Raman signal contribution returning through the inner material components is higher due to the lower lateral scattering probability of the surface components than that of the internal components. As a result, the attenuation degree of the Raman scattering signal generated by the surface components is greater than that of the inner components, enabling the detection of the Raman signals generated by internal components [22,23]. This technique was proposed by Pavel et al. in 2005 based on the study of Raman scattering photon migration theory. The spatially shifted Raman technique mitigates fluorescence interference and enables fast identification and detection of internal material components hidden under opaque media [24].

According to the introduction to the above five Raman spectroscopy techniques, their different characteristics are summarized as detailed in Table 1.

Category	Principle/ Conditions	Advantages	Disadvantages
Confocal Micro Raman Spectroscopy (CRS)	Microscopy combined with Raman spectroscopy	Precise imaging, high detection sensitivity, high spatial resolution, three-dimensional reconstruction, qualitative and quantitative analysis	Disturbed by fluorescence
Fourier Transform Raman Spectroscopy (FT-Raman)	1064 nm wavelength as laser source	Fast scanning spectra, reduced fluorescence interference	Temperature drift and specimen movement have a significant effect on the spectrum
Surface Enhanced Raman Spectroscopy (SERS)	Must be adsorbed (or close to) on metal surfaces, using gold, silver, etc., as a substrate, or using nanoparticles as a medium	Fast detection and high sensitivity	Quantitative and stability are difficult to control
Resonance Raman Spectroscopy (RRS)	When the incident light energy approaches an electron-excited state energy level	High sensitivity, high selectivity	Higher equipment cost, serious interference by fluorescence, influenced by wave number
Spatially Offset Raman Spectroscopy (SORS)	Offset a certain distance on the sample surface space	Non-destructive detection of deep biochemical composition information of samples	Optical path systems that require the excitation of light and collection of light offsets

Table 1. Comparison of the characteristics of different Raman spectroscopy techniques.

4. Application of Raman Spectroscopy in Testing Fruit and Vegetable Quality and Safety

Fruits and vegetables are rich in nutritional value and contain numerous vitamins, minerals, and fiber. The gradual development of Raman spectroscopy has resulted in significant research outcomes for internal and external quality inspection of fruits and vegetables, both domestically and internationally. Due to its specific compound spectral fingerprint characteristics recognition, it has been applied to identify and differentiate different quality parameters, such as the detection of maturity, freshness, damage, and nutrient content of fruits and vegetables. It can also quantify nutrient content in fruits and vegetables by detecting their vibrational frequencies in Raman spectra [25–31]. Furthermore, more research reports have focused on detecting pesticide residues on the surface of fruits and vegetables. Many cases have demonstrated that Raman spectroscopy is efficient for

studying pesticide and hormone residues on the surface of fruits and vegetables. It has yielded promising results in the external detection of these substances.

4.1. Application of Confocal Micro Raman Spectroscopy for Testing Fruit and Vegetable Quality and Safety

4.1.1. Quality Inspection Aspect

Qian et al. [32] proposed to discriminate fruit species by analyzing the β -carotene spectrum on the surface of fruits and using the differences in spectrum collected by the confocal Raman spectroscopy technique as the basis for discrimination. Pudney et al. [33] utilized confocal Raman micro spectroscopy to investigate the molecular states of carotenoids in tomatoes and foods, demonstrating its effectiveness in monitoring changes in their physical states. Their findings also illustrated the differentiation of the three primary carotenoids in tomatoes (lycopene, β -carotene, and lutein) and their respective physical statuses. Apart from classification and identification, confocal micro-Raman can also be employed to assess the ripeness and freshness of fruits and vegetables. Trebolazabala et al. [34] utilized a portable device and a confocal Raman spectrometer of different wavelengths to obtain spectral information on the main organic components of tomato fruits and the analysis of ripe and unripe tomatoes to identify the main organic components (carotenoids, cuticular compounds, polysaccharides, polyphenols, etc.) in the two ripening stages of this fruit, thus establishing a promising Raman signature of that distinguishes between the different components of ripe and unripe tomatoes. Monika et al. [35] used micro-Raman imaging to observe changes in the distribution of cell wall polysaccharides during the ripening of tomato fruits to discriminate the ripeness of tomatoes. Results showed ripe tomatoes contained high concentrations of pectin polysaccharides at the cell corners in thin, green walls with more uniform polysaccharide distribution. These ripe tomatoes exhibited high pectinolytic enzyme activity and increased ascorbic acid content, which reduced pectin levels. Loss of pectin during tomato fruit ripening may lead to reduced intercellular adhesion, relocation, and redistribution of polysaccharides. Sharma et al. [36] employed 532 nm laser-excited confocal micro-Raman spectroscopy to track exocarp pigmentation changes in young, early, ripe, early-ripe, mature, and post-ripe cherries and established data mining methods such as principal component analysis and hierarchical cluster analysis to analyze the spectral distribution. The study revealed that compared to the early-ripe stage, the intensity of carotenoid bands in the spectra of ripe fruits decreased as the anthocyanin bands increased. Hence, confocal micro-Raman spectroscopy exhibits great potential for noninvasive monitoring of pigment changes during sweet cherry ripening. Regarding freshness detection, Nekvapil et al. [37] confirmed that the Raman signal intensity of carotenoids in citrus fruits is an excellent indicator for identifying citrus freshness. As time passed under all conditions, the freshness of citrus samples decreased, as evidenced by a decrease in both the absolute Raman intensity signal of carotenoid bands and the relative Raman intensity. Different citrus varieties exhibited varying slopes, demonstrating the correlation between the carotenoid resonance Raman scattering signal intensity and fruit freshness. In terms of internal damage to fruits and vegetables, Chen et al. [38] conducted a study using Confocal Raman spectroscopy and chemometric methods were used to analyze the Raman spectral characteristics of intact and slightly damaged apples., The SG convolution was employed to smooth and denoise the spectral data, and the baseline correction was performed with the adaptive iterative re-addition airPLS algorithm to establish the SVM classification model. Pan et al. [39] used the laser confocal micro Raman spectroscopy (CRM) technique to collect Raman spectra of cell walls of fresh pear fruits and those infected by Alternaria alternata (A. alternata), as well as label-free in situ imaging of the chemical composition of the cell walls. The findings revealed significant signal intensity changes in the cell walls. In addition, the chemical images of the cell walls from fresh and disease-susceptible fruits were compared, and morphological structure was analyzed. The results validated the applicability of CRM for identifying changes in cell wall composition induced by fungal infections. Regarding fruit and vegetable component content analysis, Hu et al. [40] used

confocal micro-Raman spectroscopy to detect the polysaccharide content of different parts of Dendrobium. They combined with the Raman spectral information of polysaccharide components of Dendrobium, computer optimization algorithm, and chemometric method were used to screen out the spectral bands with obvious and well-specified wave peaks as the characteristic peaks of polysaccharide components to achieve the quantitative detection of polysaccharide components. Li et al. [41] demonstrated that the water content in apple and potato cells could be visualized and quantified by confocal Raman microscopy. The quantification of total and free water content was achieved based on the pull distribution images of water content and hydrogen bonding states in apple and potato tissues, and the order of water content in apple or potato cells was vesicular water > water in the cell junction region > water in the cell wall or intercellular space. The experiments demonstrated that the confocal micro-Raman technique is a precise and dependable method for investigating water content distribution and water status within apple and potato cells.

4.1.2. Security Detection Aspects

Numerous studies have shown that confocal micro-Raman spectroscopy is a valuable tool for diagnosing diseases in fruits and vegetables, identifying bacterial spoilage, and detecting pesticide residue contamination on their surfaces, which ensures food safety. Guo et al. [42] monitored the dynamics of apple infestation by the dominant spoilage bacteria. It was found that polysaccharides, cellulose, and pectin in apple cells were not uniformly distributed in the cell walls and cell interstices at different stages of infestation. The intensity of the characteristic peaks of Raman spectra gradually decreased with the increase of spoilage bacteria infestation. The results showed that Raman spectra of apples infested by bacteria at different stages displayed a clustering pattern. The discriminant model achieved an accuracy above 95% for correction and prediction sets, facilitating early detection and process analysis of rotten bacteria infestation in apples. Cai et al. [43] used confocal Raman microscopy to perform single Raman spectroscopy, multiple Raman spectroscopy, and microscopic image analysis of fresh fruits and fruits invaded by decaycausing fungi. The intensity of Raman peaks at 1008 cm^{-1} , 1154 cm^{-1} , and 1525 cm^{-1} decreased, indicating that the content of carotenoids in citrus fruit peel decreased significantly during fruit decay. Cai analyzed the critical components of fresh and spoiled fruits. The correlation coefficients of both the correction and prediction sets of the linear discriminant analysis method reached 100%, confirming its ability to effectively identify changes in fruit composition caused by spoilage bacteria. Another crucial aspect of the safety of edible fruits and vegetables is the presence of pesticide residues on their surface. Researchers have widely adopted the confocal micro-Raman technique to detect and quantify pesticide residues on the surface of fruits and vegetables. Kang et al. [44] further verified that laser micro Raman spectroscopy can be effectively applied to precisely detect pesticide residues in fruits, comparing the Raman image features on the surface of apples with and without pesticide residues and different concentrations of pesticide residues. The results indicated that the Raman image features were notably enhanced in apples with trichlorfon pesticide residues on the surface, allowing detection limits as low as 4800 mg/kg. Sun et al. [45] used confocal Raman spectroscopy to acquire the spectral image information of apples and S-G17 times smoothing to reduce the instrument noise interference and shape-matching background removal method to remove the fluorescence background interference. The results indicated that the Raman signal of the phosphorothioate group at 632 cm $^{-1}$ could be used to identify the chlorpyrifos pesticide residues on the surface of apples, with a detection limit of 48 mg/kg. The related studies have been summarized in Table 2.

Classification	Analytes	Raw Material	Measurement Technique	Result	References
Quality inspection aspect	physical states	Tomato	CRS	Successfully confirmed	Pudney et al., 2011 [33]
Quality inspection aspect	maturity	Tomato	CRS	Successfully confirmed	Trebolazabala et al., 2013 [34]
Quality inspection aspect	maturity	Tomato	CRS	Successfully confirmed	Monika et al., 2017 [35]
Quality inspection aspect	maturity	Cherry	CRS	Successfully confirmed	Sharma et al., 2022 [36]
Quality inspection aspect	Freshness	Citrus fruits	CRS	Successfully confirmed	Nekvapil et al., 2018 [37]
Quality inspection aspect	Distinguish damaged apples Distinguish	Apple	CRS	classification accuracy: 97.8%	Chen et al., 2018 [38]
Quality inspection aspect	between fresh pear fruits and those infected by Alternaria alternata (A. alternata)	Pears	CRS	Success differentiation	Pan et al., 2017 [39]
Quality inspection aspect	Analysis of polysaccharide content	Dendrobium	CRS	Successfully confirmed	Hu et al., 2017 [40]
Quality inspection aspect	Quantitatively determine water content and hydrogen bond status in fruit and vegetable cells.	Apple, Potato	CRS	The water contents in apple or potato cells followed the order: vacuole water > water in cell junction regions > water in the cell wall or intercellular spaces	Li et al., 2022 [41]
Security detection aspects	Distinguish apples infected with bacteria at different stages	Apple	CRS	classification accuracy: 97.8%	Guo et al., 2022 [42]
Security detection aspects	Distinguish between fresh and spoiled fruits	Citrus fruit	CRS	classification accuracy $\approx 100\%$	Cai et al., 2023 [43]
Security detection aspects	Analysis of trichlorfon pesticide residues on apple surfaces	Apple	CRS	Detection limit: 4800 mg/kg	Kang et al., 2021 [44]

Table 2. Confocal micro-Raman spectroscopy is applied to detect fruit and vegetable quality and safety rapidly.

4.2. Fourier Transform Raman Spectroscopy for Testing the Quality and Safety of Fruits and Vegetables

4.2.1. Quality Inspection Aspect

Muik et al. [46] demonstrated that Fourier transforms Raman spectroscopy combined with pattern recognition can be used to identify and distinguish olives having different characteristics related to their quality conditions, including intact olives, olives with frostbite, olives collected from the ground, fermented olives, and olives with the disease, based on data preprocessing of the obtained spectral data and analysis by chemometric methods (SIMCA method). The study achieved high predictive power for each category, with 93% for intact olives, 93% for frostbite, 96% for ground-collected, and 92% for fermented olives. However, the sample size for diseased olives was too small to determine a predictive category. Overall, this method provides a reliable way to select good-quality olives. Rubayiza was the first to utilize Fourier Transform Raman Spectroscopy for coffee analysis [47]. The visual examination of the Raman spectra of the lipid fraction extracted from *Coffea arabica* (Arabica) and *Coffea canephora* (robusta), and *Coffea liberica* (liberica) samples show differences in the mid-wavenumbers' region. Specifically, the spectrum of Arabica presents two scattering bands at 1567 and 1478 cm⁻¹, which are absent in the spectra of Robusta and Liberica. Raman spectroscopy can be utilized to differentiate between arabica coffee samples with high and low contents of kahweol.

Pierna et al. [48] demonstrated that the Fourier transform Raman spectroscopy technique combined with chemometric methods like partial least squares discrimination or support vector machine (SVM) techniques can be used as a fast and reliable method to identify the origin of honey. The accuracy rates of all models were found to be between 85% and 90%. Yu et al. [49] measured the unsaturation values of carbon-carbon double bonds using Fourier transform Raman spectroscopy to distinguish common edible vegetable oils, as the carbon-carbon double bond content in the fatty acids of different edible vegetable oils varies. The research results showed that the integrated value of the absorption peak area at 1747 cm⁻¹ was used as a reference, and a particular distinction between different types of common edible vegetable oils was achieved by comparing the integrated value of the absorption peak area at 1654 cm^{-1} . Stawoska et al. [50] used FT-Raman spectroscopy to analyze endosperm and alcohol-soluble proteins extracted from wheat embryos. The researchers conducted a quantitative analysis of the results and found that alcohol-soluble protein complexes in wheat could interact with other proteins through weak, low-energy hydrogen bonds. This new finding explains how wheat alcohol-soluble protein complexes impact the secondary structure of the remaining alcohol-soluble protein. Yang et al. [51] proposed a rapid spectroscopic method for determining vitamin C in food and pharmaceutical products using infrared and Raman techniques was proposed. In this study, FT-Raman spectroscopy was used in conjunction with partial least squares (PLS) regression to quantify vitamin C in powdered mixtures and solutions. The results indicated a high correlation in the prediction of the adopted methods. R^2 values were 0.95 for FT-Raman, with an overall prediction error of 0.2–3.0%. Raman techniques can quantify vitamin C in foods and pharmaceutical products. Batsoulis et al. [52] developed a method for determining fructose and glucose in honey using Fourier transform Raman spectroscopy and spectrometer software. The fructose content ranged from 24.0% to 10.8%, and the glucose content ranged from 21.1% to 32.2%. Baranska et al. [53] used Fourier transform Raman spectroscopy to determine the lycopene and β -carotene contents in tomato fruits and related products. The FT-Raman spectra of tomato paste showed three strong bands at 1510 cm⁻¹, 1156 cm⁻¹, and 1005 cm^{-1} . It has been found that FT-Raman spectroscopy can be successfully applied for the identification of carotenoids directly in plant tissue and food products without any preliminary sample preparation. Agnieszka et al. [54] used Fourier transform Raman spectroscopy and thermogravimetric analysis to investigate the changes in gluten protein structure and its thermal properties induced by seven commercial dietary fibers (fruits, vegetables, and cereals), respectively. Bread dough with a consistency of 500 Farinograph Units (FU) was prepared using a blend of wheat starch and wheat gluten, with a dietary fiber ranging from 3 to 18% w/w. The results showed that all dietary fibers, except oats, caused similar changes in the secondary structure of gluten proteins.

4.2.2. Security Detection Aspects

Zhou et al. [55] conducted Raman tests on common fruits and pesticides using a Fourier transform Raman spectroscopy and obtained corresponding characteristic Raman spectra of fruits and pesticides. The tests conducted on the surface of fruits with pesticides showed that the Raman spectra could simultaneously show the characteristic spectra of fruits and pesticides to identify various pesticides on the surface of fruits. The experimental results show that the Fourier transform spectroscopy technique can rapidly and effectively detect the existence of pesticide residues on fruit surfaces. The related studies have been summarized in Table 3.

Classification	Analytes	Raw Material	Measurement Technique	Result	References
Quality inspection aspect	Distinguish olives of different qualities	Olive	FT-Raman	Successfully confirmed	Muik et al., 2004 [46]
Quality inspection aspect	discrimination of Arabica and Robusta coffees	Coffee	FT-Raman	Successful differentiation	Rubayiza et al., 2005 [47].
Quality inspection aspect	Identify the source of honey Distinguish	Honey	FT-Raman	classification accuracy: 85–90%	Pierna et al., 2011 [48]
Quality inspection aspect	different types of common edible	Edible vegetable oil	FT-Raman	Successful differentiation	Yu et al., 2012 [49]
Quality inspection aspect	Quantitative Analysis of Prolamin	Wheat	FT-Raman	Alcohol-soluble protein complexes in wheat could interact with other proteins through weak, low-energy hydrogen bonds.	Stawoska et al., 2021 [50]
Quality inspection aspect	Identification of vitamin C content in foods and medicines.	Foods, Medicines	FT-Raman	R ² : 0.95	Yang et al., 2002 [51]
Quality inspection aspect	Quantitative determination of fructose and glucose in honey	Honey	FT-Raman	fructose content: 24.0–10.8%; Glucose content: 21.1–32.2%	Batsoulis et al., 2005 [52]
Quality inspection aspect	Identification of lycopene and β-carotene content	Tomato fruits, Related product	FT-Raman	R ² = 0.91 for lycopene; R ² = 0.89 for β-carotene.	Baranska et al., 2006 [53]
Quality inspection aspect	Identification of gluten structure and its thermal properties caused by dietary fibers	Fruits, Vegetables, Grains	FT-Raman	Successfully confirmed	Agnieszka et al., 2016 [54]
Security detection aspects	Detection of pesticide residues on the surface of fruits	fruit	FT-Raman	Successfully confirmed	Zhou et al., 2004 [55]

Table 3. Application of Fourier transform Raman spectroscopy in rapid detection of fruit and vegetable quality and safety.

4.3. Surface-Enhanced Raman Spectroscopy for the Assessment of the Quality and Safety of Fruits and Vegetables

4.3.1. Quality Inspection Aspect

Gopal et al. [56] employed novel and simple nanocomposites (silver and gold nanoparticles on graphene nanosheets, Ag@G and Au@G) as a substrate for SERS combined with the Raman technique to increase the Raman signal to conduct a study of fresh fruits and vegetables before and after refrigeration for up to 2 weeks, thus assessing the freshness of fruits and vegetables based on the changes in their surface Raman spectra. The results showed that the Raman spectra changed significantly with decreasing fruit freshness and increasing refrigeration time. This indicates that the technique can successfully distinguish between fresh and stored fruits and vegetables in the market and that the detection method is simple, sensitive, rapid, and non-destructive.

4.3.2. Security Detection Aspects

SERS technology has been extensively applied for detecting pesticide residues in fruits and vegetables due to its capability of detecting trace analytes. There are various pesticide residues involved in fruits and vegetables, such as triazophos residues in the flesh of navel oranges [57], mixed pesticide residues in navel orange peel [58], the pesticide residues of killing thion in watermelon [59], the pesticide residues in rapeseed oil [60] organic pesticide residues in yam [61], etc. Liu et al. [62] used surface-enhanced Raman spectroscopy technology to investigate the presence of organophosphate pesticides, imidacloprid, and chlorpyrifos, in the residues of navel orange peel. The gold gel was utilized as an enhancement substrate to capture the spectra of navel oranges. The obtained spectral data were preprocessed using the first-order derivative method and analyzed by the PLS algorithm. A highly reliable quantitative prediction model was established with $R_P = 0.909$, RMSEP = 3.338 mg-L⁻¹. Based on the comparative differences in characteristic peaks of pesticide Raman spectroscopy, the experimental results demonstrate that mixed pesticide residues on navel orange peel can be analyzed qualitatively and quantitatively. Yang et al. [63] proposed an improved probability of detection (POD) method based on surface-enhanced Raman spectroscopy for quantitatively analyzing carbendazim pesticides. The method focused on the rapid screening of pollutants in fruits. The results indicated a 100% detection rate at concentrations of not less than 1 mg/kg, with a detection limit of carbendazim at 0.5 mg/kg. The false favorable rates for the semiquantitative analysis method were 0% and 5% at 0.5 mg/kg and 2.5 mg/kg, respectively. The experiment has validated the efficacy of this method in meeting the requirements of swift on-site detection. Chen et al. [64] verified the effectiveness of surface-enhanced Raman spectroscopy technology in the rapid on-site detection of carbendazim in fruits and vegetables by carrying out pre-optimization experiments and on-machine testing conditions. Six Raman characteristic shifts of carbendazim were identified, and a POD model was established to detect it in fruits like apples and citrus fruits at a limit of 0.5 mg/kg and in vegetables like chilli peppers at a limit of 1.0 mg/kg, from the perspective of detection probability. The related studies have been summarized in Table 4.

Table 4. Application of surface-enhanced Raman spectroscopy in rapidly detecting fruit and vegetable quality and safety.

Classification	Analytes	Raw Material	Measurement Technique	Result	References
Quality inspection aspect	Freshness	Fruits, Vegetables	SERS	Successfully confirmed	Gopal et al., 2016 [56]
Security detection aspects	Triazophos residues	Orange	SERS	Successfully confirmed	Wang et al., 2016 [57]
Security detection aspects	Pesticide residues of killing thion	Watermelon	SERS	Successfully confirmed	Chen et al., 2018 [59]
Security detection aspects	Fenitrothion pesticide residues	Rapeseed oil	SERS	Successfully confirmed	Yang et al., 2019 [60]
Security detection aspects	Dimethoate pesticide residues	Yam	SERS	Successfully confirmed	Liu et al., 2022 [61]
Security detection aspects	Organophosphorus pesticide imifos and Chlorpyrifos residue	Orange	SERS	Successfully confirmed	Liu et al., 2018 [62]
Security detection aspects	carbendazim pesticides	Fruit	SERS	Successfully confirmed	Yang et al., 2002 [63]
Security detection aspects	carbendazim pesticides	Fruit, Vegetable	SERS	The detection limit of apples and oranges: 0.5 mg/kg; The detection limit of chili peppers: 1.0 mg/kg.	Chen et al., 2022 [64]

4.4. Resonance Raman Spectroscopy Can Be Applied to Testing the Quality and Safety of Fruits and Vegetables

4.4.1. Quality Inspection Aspect

Resonance Raman spectroscopy effectively detects the components of fruits and vegetables, specifically carotenoids, and their impact on the quality of these foods and the correlation between their contents. Badgujar et al. [65] used resonance Raman spectroscopy to investigate the regulation and accumulation of carotenoids in various plant and animal systems and to identify carotenoids with high sensitivity and specificity without needing pre-processing or extraction. The resonance Raman spectroscopy method successfully identifies the occurrence and changes of carotenoids during their growth. It can be applied to identifying carotenoids in all plants without any sample pretreatment. The resonance Raman spectroscopy (RRS) model achieved legal validity and proved reliable in assessing the carotenoid status and fruit and vegetable intake. Gonzálvez et al. [66] used transmission Resonance Raman Spectroscopy to monitor the LED-induced increase in the carotenoid content of grapes. The obtained experimental data demonstrated an approximately 5-fold increase in carotenoid content compared to the original control samples. Bhosale et al. [67] investigated the determination of carotenoid content in various agricultural products and fruit juices by resonance Raman spectroscopy. They compared it with the results obtained from the extraction and high-pressure liquid chromatography detection methods. The study showed a strong correlation between the carotenoid content obtained by these two methods, specifically in detecting fruit juices and the same type of agricultural products at the same maturity stage. There are also relevant experiments that can be based on detecting changes in the spectral characteristics of carotenoids on the surface of watermelon fruit that can be used as a marker for monitoring fruit ripeness [68]. Using partial least squares discriminant analysis and resonance Raman spectroscopy, the pre-processed spectra were analyzed to identify the different ripeness of four watermelon varieties achieving an accuracy of over 85%. The study confirms that the resonance Raman spectral signal can effectively monitor the changes of surface carotenoids during watermelon ripening, and it can be utilized as an indicator for monitoring nutritional quality and ripeness. In addition, Braeuer et al. [69] investigated the feasibility of online monitoring of CO₂-assisted drying of two fruits, mango and persimmon, using resonance Raman spectroscopy, thus concluding that when pressure is released from the drying vessel at the end of the CO_2 drying process, a large amount of water spills out of the fruit slices together with CO_2 and the absorption of CO_2 in the fruit matrix facilitates the extraction of water.

4.4.2. Security Detection Aspects

Regarding safety detection, Ranjan et al. [70] proposed a gold nanoparticle-enhanced resonance Raman spectroscopy technique to detect trace concentrations of pesticides. This technique uses gold nanoparticle-enhanced Raman spectroscopy signals to observe the corresponding characteristic peaks for detecting low concentrations of pesticides of about 10^{-6} mol/L. The study demonstrated that tiny pesticides could be detected rapidly in oranges. The related studies have been summarized in Table 5.

Table 5. Application of resonance Raman spectroscopy in rapid detection of fruit and vegetable quality and safety.

Classification	Analytes	Raw Material	Measurement Technique	Result	References
Quality inspection aspect	Monitoring LED-induced carotenoid increase in grapes	grapes	RRS	Successfully confirmed	Gonzálvez et al., 2013 [66]
Quality inspection aspect	maturity	Watermelon	RRS	accuracy: $\geq 85\%$	Dhanani et al., 2022 [68].

Classification	Analytes	Raw Material	Measurement Technique	Result	References
Quality inspection aspect	The feasibility of applying in situ Raman spectroscopy for the online monitoring of the supercritical carbon dioxide (SC–CO ₂) drying of fruits	mango, persimmon	RRS	The feasibility of applying in situ Raman spectroscopy for the online monitoring of the supercritical carbon dioxide (SC-CO ₂) drying of fruits	Braeuer et al., 2017 [69]
Security detection aspects	Trace pesticide residues	Orange	RRS	Detection limit: 10 ⁻⁶ mol/L	Ranjan et al., 2016 [70]

Table 5. Cont.

4.5. The Utilization of Spatially Offset Raman Spectroscopy for Testing the Quality and Safety of Fruits and Vegetables

4.5.1. Quality Inspection Aspect

Spatially offset Raman spectroscopy technology has been applied to detect the quality aspects such as fruit and vegetable classification and maturity. Morey et al. [71] examined potatoes as their research subject to investigate whether the varying nutrient proportions could serve as an identifier of potato varieties and their geographical origins. The PLS-DA model established using Raman spectra collected from various potato varieties exhibited a 100% prediction accuracy. The PLS-DA, developed based on the variations in nutrient composition of potatoes between two regions, provides a high prediction accuracy of 84.3–90.9% and thus can effectively distinguish the origin of potatoes. Qin et al. [72] evaluated the internal ripeness of tomatoes based on nondestructive testing by spatially offset Raman spectroscopy. The inhomogeneous generation of carotenoid content during tomato ripening was adopted as an assessment criterion. The Raman spectral information of carotenoids from tomatoes at different ripening stages was collected using Spatially Offset Raman Spectroscopy with Self-Modeling Mixture Analysis (SORS-SMA) method. The Raman peaks were evaluated by calculating the spectral information scatter (SID) using pure lycopene as a reference. Results showed that the SID values decreased with the maturity of tomatoes, effectively distinguishing the maturity of tomatoes based on the rise and fall of SID values.

4.5.2. Security Detection Aspects

Farber et al. [73] verified that spatial offset Raman spectroscopy technology can rapidly and non-destructively detect Zebra chip disease (ZC) and Potato virus Y diseases (PVY) in potatoes, based on Raman identification of chemical changes in tubers associated with ZC or PVY. Chemometric analysis of spectra from intact potato tubers demonstrated a ZC detection accuracy of over 90%. Moreover, potatoes from plants with either ZC or PVY diseases can be distinguished from each other and healthy tubers with an accuracy of approximately 95%. The related studies have been summarized in Table 6.

Table 6. Application of spatially offset Raman spectroscopy in rapidly detecting fruit and vegetable quality and safety.

Classification	Analytes	Raw Material	Measurement Technique	Result	References
Quality inspection aspect	Distinguish the origin of potatoes	Potato	SORS	Prediction accuracy: 84.3–90.9%	Morey et al., 2020 [71]
Quality inspection aspect	maturity	Tomato	SORS	Successfully confirmed	Qin et al., 2012 [72]

Classification	Analytes	Raw Material	Measurement Technique	Result	References
Security detection aspects	Detection of Zebra chip disease (ZC), Potato virus Y diseases (PVY) and healthy tubers in potatoes.	Potato	SORS	Prediction accuracy: 95%	Farber et al., 2020 [73]

Table 6. Cont.

5. Comparison of Raman Spectroscopy with Other Intelligent Non-Destructive Techniques

Nondestructive testing techniques have been extensively employed in fruits and vegetables. In addition to the Raman spectroscopy techniques discussed in this paper, other techniques such as near-infrared spectroscopy, RGB vision detection, electronic nose inspection, acoustic characteristics detection, and dielectric properties detection have also been reported in research [7]. Each of these testing techniques possesses unique advantages in assessing the quality and safety of fruits and vegetables. There is potential for developing more targeted nondestructive testing methods that cater to specific types of products and capitalize on the strengths of the abovementioned techniques. Establishing standard specifications will further advance the practicality and utilization of nondestructive testing methods.

5.1. Near-Infrared Spectroscopy

Near-infrared spectroscopy is an absorption spectroscopy technique that irradiates a sample with near-infrared light. This causes molecular vibrations of the substance to elevate from a ground state to an excited state. By examining the absorption spectra produced in the near-infrared region, the characteristic information of organic molecules and their hydrogen-containing groups, based on the X-H vibrations (where X = C, N, O) and their respective multiplications and ensemble frequencies can be determined [74,75]. The near-infrared spectral region exhibits distinct spectral absorption peaks for most structural components of fruits and vegetables. Utilizing the discerning data from the near-infrared spectra, it is primarily employed for identifying fruit and vegetable species and detecting indicators of internal quality such as maturity, freshness, and content [76–81]. In terms of signal reception, NIR outperforms Raman. However, the Raman spectroscopy technique provides more precise Raman spectra, making spectrum analysis more convenient and making spectrum analysis more convenient. NIR spectroscopy can qualitatively and quantitatively analyze multiple components of fruits and vegetables simultaneously due to its high resolution. However, low molecular vibration absorption and sensitivity render it unsuitable for analyzing trace components.

In contrast, by combining microscopic techniques, Raman spectroscopy can enhance detection accuracy and trace component analysis. Furthermore, near-infrared radiation is highly absorbed by water molecules and greatly influenced by sample moisture when testing the quality of fruits and vegetables. On the other hand, Raman spectroscopy light can effectively penetrate through water and detect the quality of fruits and vegetables without interference. In comparison, the Raman spectroscopy technique is more suitable for detecting trace components in fruits and vegetables and can be used for detecting through aqueous solutions.

5.2. RGB Vision Detection Technology

RGB vision detection technology simulates human vision to observe and judge objects. Its main principle involves using image acquisition equipment to detect objects through image signals transmitted to a computer. The obtained image information is then processed and analyzed for the automatic identification, classification, and detection of objects [82]. RGB vision detection technology is widely used to classify and identify external quality

characteristics of fruits and vegetables, including size, shape, color, texture, and appearance defects [82–86]. RGB vision detection technology provides highly reliable and precise analysis of fruit and vegetable sample images, primarily focusing on appearance inspection. However, limitations exist in its ability to analyze the qualitative and quantitative aspects of fruit and vegetable composition quality. Raman spectroscopy technology, with its unique characteristic fingerprint spectra and the ability to detect the extent of absorption changes in the spectrum caused by fruits and vegetables, has the potential to acquire chemical information, including quantitative and qualitative analysis of their components. Therefore, Raman spectroscopy technology surpasses other methods in the qualitative and quantitative and quantitative and qualitative and qualitative and quantitative and qualitative and spectroscopy technology surpasses other methods in the qualitative and quantitative detection of internal quality in fruits and vegetables.

5.3. Electronic Nose Detection Technology

Electronic nose detection technology is a sensor technology based on pattern recognition, which is mainly a combination of three systems: sensor array, signal pre-processing, and pattern recognition. The working principle of electronic nose detection technology is based on the simulated mechanism of animal olfactory perception. The chemical composition of the measured gas undergoes a chemical reaction with the sensor array, which converts the chemical response into an electrical signal. The sensor arrays have different sensitivities to the measured gas, generating a response spectrum for fast and accurate qualitative or quantitative detection of gas components [87]. Electronic nose detection technology can rapidly and accurately detect the qualitative or quantitative compositions of gases. This technology can detect various odor components emitted by different types of fruits and vegetables, enabling the identification of their varieties and evaluating their quality. In addition, electronic nose detection technology can predict their storage life by analyzing the changes in the content of volatile organic substances in fruits and vegetables [88–90]. Electronic nose technology is susceptible to changes in external environmental factors, such as temperature, humidity, and odor, which makes it necessary to strictly control the sample detection environment. Additionally, the stability of the sensor is inadequate, which may pose challenges in accurately recognizing objects during the detection process. Raman spectroscopy technology is unaffected by sample volatility, thereby removing the requirement for strict control of the detection environment. Electronic nose technology detects the general information of volatile components of fruits and vegetables by sensing their surface odor. However, to enable the detection of these volatile substances by electronic nose sensors, the samples require pre-processing techniques such as heating, stirring, or other techniques. In contrast, Raman spectroscopy is a non-destructive analytical technique that enables direct spectral measurements of samples without any physical or chemical treatment.

5.4. Acoustic Signature Detection Technology

The acoustic feature detection technology generates acoustic waves by tapping or subjecting fruits and vegetables to external pressure, resulting in an acoustic resonance phenomenon. Due to differences in the internal structure and physical properties of fruits and vegetables, the resonant frequencies of the absorbed sound waves are different, thus analyzing the frequency, amplitude, time, and other characteristic signals of fruits and vegetables to assess and identify the properties of fruits and vegetables [91]. An acoustic characterization is a valuable tool for assessing the quality of fruits and vegetables, particularly their ripeness, freshness, texture, and flavor [92–95]. Acoustic feature detection technology can analyze the physical properties of fruits and vegetables, providing limited insights into their internal quality. However, its analysis of chemical composition is relatively limited. In contrast, Raman spectroscopy can determine samples' chemical composition and molecular structure by measuring their vibrational and rotational modes. Additionally, acoustic feature technology cannot ensure the preservation of fruits and vegetables during impact or tapping detection, as it may result in deformation, injury, or even destruction of the samples. On the other hand, Raman spectroscopy analyzes the

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sample by scattering specific wavelengths of light when exposed to an excitation light source. The acoustic feature is a promising non-invasive approach for assessing the quality of fruits and vegetables, as it does not require physical interference with the sample, which allows for the preservation of sample integrity and natural fruit and vegetable quality.

5.5. Dielectric Properties Testing Technology

Dielectric properties detection technology is primarily utilized to measure the dielectric properties of fruits and vegetables under the action of high-frequency electric fields, particularly the dielectric constant, to analyze and detect the nature and characteristics of the substance through changes in the dielectric constant [96,97]. The dielectric properties of fruits and vegetables have been utilized to obtain data on their dielectric constant and electrical conductivity for fruit and vegetable testing. Quality indices, including water content, maturity, and hardness, have been inferred from this data. Such techniques allow for the discrimination and sorting of fruit and vegetable ripeness based on the aforementioned indices. The data can be utilized to assess the maturity and quality of fruits and vegetables [98–102]. The dielectric characteristics of fruits and vegetables are affected by many factors, such as the frequency and temperature of the electrical measurement interference test, resulting in the unstable acquisition of the dielectric characteristics signal. In contrast, Raman spectroscopy is an optical analysis method that captures signals by using laser beams interacting with the sample without any direct contact or the need for an external electric field, which can stably acquire the signal of fruit and vegetable samples. Dielectric property detection technology poses challenges in detecting subtle changes in trace components within fruits and vegetables. However, Raman spectroscopy technology has a high resolution, which can detect trace differences in fruits and vegetables and perform sensitive detection and evaluation of trace molecular components.

6. Conclusions and Outlook

The application of Raman spectroscopy in fruit and vegetable quality detection is promising. However, it is still a relatively new fruit and vegetable detection technology. Many technical and application difficulties must be solved, such as spectral fluorescence interference, sensitivity, enhanced Raman signal, equipment cost and miniaturization. In addition, Raman spectroscopy requires establishing a reliable spectral database to rapidly search and identify chemical components in fruits and vegetables.

Most fruits naturally possess fluorescence, making the detection highly susceptible to fluorescence signals and thus making it challenging to collect high-intensity Raman signals. Additionally, instrument noise interference may also impact the detection results. The processing methods are non-uniform. Therefore, we explore more effective spectral preprocessing methods. Since the Raman spectral signal is weak, selecting the appropriate laser wavelength is essential to obtain the optimal Raman signal. Additionally, by developing new substrates, researchers can improve the stability and sensitivity of Raman technology and expand its application in the study of molecular Raman signals from analyte substances.

Cost reduction and increased convenience of equipment are the future development trends. Raman spectroscopy instruments are mainly used for laboratory science and technology research, but their high prices pose a challenge. Furthermore, the applications of fruit detection are diverse, and detecting equipment, whether portable or part of a production line, needs to be optimized and further developed to achieve low cost, simple operation, and low susceptibility to interference, among other factors, in response to changes in demand and technological advancements. These optimizations will promote the applicability of Raman spectroscopy technology and facilitate its further development and application.

The Raman spectroscopy's "fingerprint spectrum" feature enables the characterization of the composition and content information of various substances in fruits and vegetables without the need for sample pretreatment, which provides a rapid analysis method for fruit and vegetable detection. Further studies are needed to investigate the nutritional structure of fruits and vegetables using Raman spectroscopy. In addition, existing Raman spectroscopy techniques have mainly been applied to thin-skinned produce, and research on thicker and harder-skinned produce needs to be improved.

In conclusion, Raman spectroscopy is undergoing rapid development and holds great potential for broader application in the quality inspection of fruits and vegetables. As a novel, rapid, and non-destructive testing technology, it can address diverse measurement requirements in both laboratory and field settings. As a result, this technology can enhance the quality and safety of fruits and vegetables, providing consumers with better quality assurance.

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