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Terahertz Spectroscopy in the Field of Food Safety: A Review

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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Review Article

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ABSTRACT

with the continuous improvement of people's living standards, food quality and safety problem has caused the wide attention of the whole society, the traditional food quality detection method has certain limitations, terahertz spectral technology (THZ) is a new spectral detection technology, its strong perspective, efficient spectral resolution ability, special fingerprint and nondestructive, in the field of food quality and safety detection has important application value. Due to the high efficiency of food quality and the need for accurate nondestructive testing, automated nondestructive testing for food quality and safety has aroused great interest from the food sector. Faster, accurate, reliable and simplified techniques are needed to classify good quality. With their unique low-energy, penetrating, rapid and non-destructive techniques are widely used as a non-destructive and cost-effective method in the food sector. This paper summarizes the principles and instruments of terahertz spectrum, and introduces the research progress of terahertz spectral detection in food, analyzes the problems in the field of food quality and safety detection, and discusses the future development trend of terahertz spectral technology.

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1. INTRODUCTION

Due to the rapid development of the society, people begin to pursue a higher quality of life. Consumer awareness and demand for quality food make the safety monitoring and quality assessment of the food industry develop towards a reliable, accurate and rapid method [1]. The quality evaluation process in some food industries is still manual by trained personnel, which is a tedious, time-consuming, expensive and complex process. Many precise but destructive methods have been invented to obtain the quality of the produce, but the subjective methods are very easy to go wrong. On the other hand, many other quality detection methods require professionals to use chemical reagents to extract and separate, preprocess, complicated operation, complicated sample preparation, excessive cost and destroy the samples. Therefore, automated non-destructive quality assessment methods are gaining popularity by reducing waste, improving efficiency and improving quality assessment techniques [2]. Research to develop nondestructive quality evaluation methods is ongoing. Among them, spectroscopy and imaging are the two main optical technologies, with the application of [3-6] in food safety and quality detection.

Spectroscopy (ultraviolet, near-infrared, MIR, visible light and Raman chemical imaging), isotope analysis, chromatography, thermal analysis have been successfully applied in the food industry. However, as an emerging technology, THz has its unique characteristics compared with NIR. IR are less tolerant to irregularly shaped foods and are prone to scattering effects, making them unsuitable for sensing application [7].

Terahertz (THz) waves are located in the frequency range of the electromagnetic spectrum between 0.1-10THz (wavelength 3mm-30μ m), located between infrared and microwave wavelengths, and in a special position. It has the dual nature of electronics and photonics. Therefore, Terahertz spectroscopy has some unique properties compared with other spectroscopy technologies. Traditional methods such as X-ray and microwave may not be able to provide some information about sensing, and Terahertz can be used to obtain such information [8]. THz technology may be used for food safety detection due to its transience, high permeability, wide band, coherence and low functionality. In recent years, the rapid development of semiconductor and ultrafast laser technology has solved the problem of terahertz radiation source and detector, and promoted the application of THz technology. Among them, it has broad application prospects in biomedicine [9], national defense [10], remote sensing [11], security [12]. In recent years, researchers have found that the high absorption of polar materials (such as water, ethanol and sugar) makes it possible to use polar materials in quantitative solutions [13-14]. Second, it can penetrate many common physical barriers, such as packaging materials, which allows terahertz waves to be used for defect identification purposes. THz waves can be used to identify defects in finished products for real-time production applications [15]. Terahertz technology has gradually demonstrated its potential and advantages in the field of food safety. From the point of view of food safety nondestructive testing, this paper introduces the principle of THz spectroscopy, reviews the achievements of THz technology in food safety field, and summarizes the application of THz technology in harmful residues, including foreign residues, antibiotic residues, mycotoxin residues and pesticide residues.

2. TERAHERTZ SPECTROSCOPIC TECHNIQUES

Terahertz spectroscopy is a technique for material analysis and detection using electromagnetic waves in the terahertz (THz) band (frequency range usually between 0.1 and 10 THz). THz wave is between infrared and microwave, with coherence, absorption, low energy, nondestructive, transient state, high resolution, suitable for non-destructive detection and material characterization [16].

2.1 Principle of Terahertz Time-domain Spectroscopy Techniques

The terahertz wave frequency range is 0.1-10 THz, with a wavelength range of 0.03-3 mm. This frequency band lies between microwave (MW) and infrared (IR) radiation in the electromagnetic spectrum, forming a bridge between the two. It is located in the middle region of electronics and photonics [17,18], and the superior characteristics of both light and electricity make up for the "terahertz gap" [19], which plays an important role in the study of material absorption characteristics. Before the 1990s, due to the technical bottleneck of terahertz source and detection technology, the terahertz frequency band has not been fully developed and utilized. With the technological breakthrough of femtosecond laser and the rapid development of high-speed photoconductive materials, terahertz technology has ushered in a large range of application and become a new research hotspot in the 21st century. Within this frequency band, many naturally occurring substances exhibit different absorption characteristics, forming unique absorption peaks that can be regarded as the "fingerprint" spectrum of matter. Terahertz spectroscopy is very sensitive to the vibration, rotational and vibrational energies of biomolecules, because the vibration frequencies of these molecules are exactly within the THz frequency range. Terahertz spectroscopy uses matter to measure the absorption, transmission and scattering of terahertz radiation to obtain information about matter. Different substances have different molecular structures and chemical components, so their absorption characteristics in the terahertz spectra will also be different. This technology has wide applications in biomedicine, materials science and safety testing, and can be used in the identification and detection of biomolecules, drug development, pathological tissue diagnosis and non-invasive biological imaging. Overall, THz spectroscopy is able to provide the fingerprint characteristics of matter, thus helping us to gain insight into the structure, composition, and properties of matter.

2.2 Generation of THz Waves

In terms of the research category, the theory and technology of microwave have been mature in recent years, and the infrared theory and technology are gradually being improved, and there is still a big gap in the technical application and theoretical exploration of terahertz band. With the development of technology, researchers have found that different THz excitation modes have different electromagnetic wave bandwidth. At present, the optical methods that can stimulate broadband THz wave mainly include photoconductive antenna technology and optical rectification effect.

The basic principle of photoconductive antenna technology is that the photoconductive material can excite electrons and generate electron-pore pairs. Then, under the action of an applied electric field, electrons and holes move to form a current, which realizes the optical signal to an electrical signal. Photoelectric conductivity material characteristic is very important to the performance of radiation source, among them, the low temperature growth of gallium arsenide (LT-GaAs), indium phosphate (InP) and defective silicon (Si) wafer commonly used materials, its gap width, carrier mobility and life, the generation of terahertz radiation efficiency and spectrum characteristics has a direct and key role.

Optical rectification effect Photorectification radiation is a widely used technology in the field of terahertz wave generation, whose core is the nonlinear optical effect. Light produces electron-pore pairs on the photorectifying material, and the nonlinear behavior of the material causes the current to flow in only one direction, and the light intensity changes the current size and direction, thus realizing signal modulation.

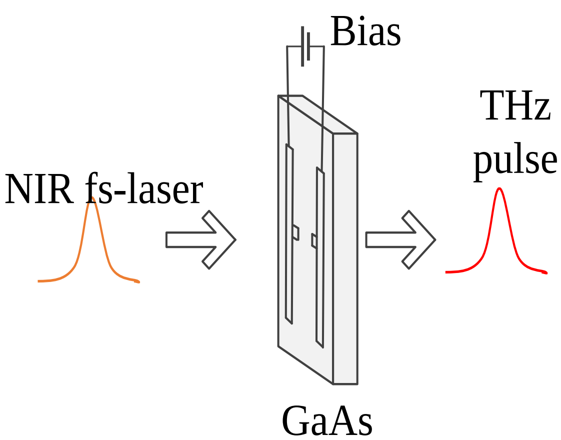

Fig. 1. Schematic diagram of terahertz radiation generated by photoconductive antenna

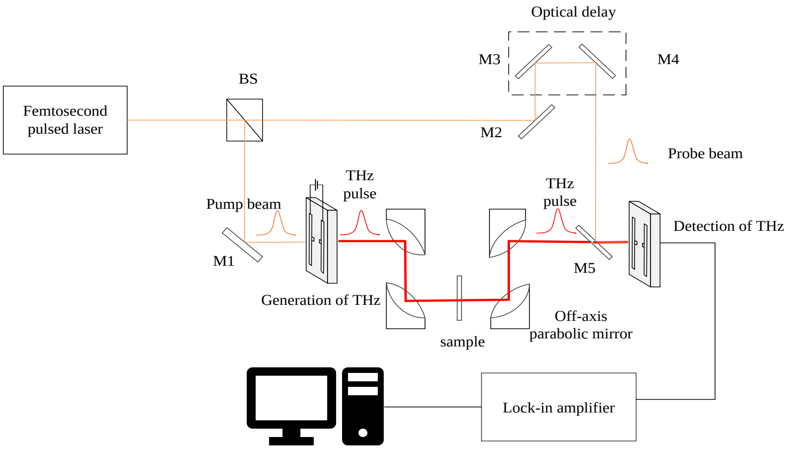

Fig. 2. Structure diagram of terahertz time domain spectroscopy system

Compared with the photoconductive antenna technology, the photorectification method does not require the action of forbidden band transition and external electric field, and the device is relatively simple and does not require a complex electrode structure. In addition, the optical rectification method can withstand higher pump power while the hardware equipment is not damaged, and has high scalability. However, the sensitivity of the optical rectification effect is low, which may not detect very weak signals, and can only work in a specific frequency range, which is not suitable for the application scenarios of wide band.

2.3 Principle of the THz Time-domain Spectral System

The THz time domain spectral sampling system is a typical pulse wave system. A schematic structure of the THz temporal spectral sampling system is shown in Fig. 2. The terahertz time domain spectrum system consists of femtosecond ultrafast laser emitter (Femtosecond pulsed laser), terahertz wave emitter (Generation of THz), terahertz wave receiver (Detection of THz), time delay controller (Optical delay) and other modules. The basic principle is that the femtosecond laser pulse emitted by the pump laser is divided by the spectrolens to form two different beams. All the way, it directly acts on the photoconductive material gallium arsenide, thus stimulating the electromagnetic pulse in the terahertz frequency band. The other route, as a detection light, is adjusted by the optical delay element, converges with the terahertz pulse in the collinear path, and is detected through the terahertz detection element. To achieve the precise regulation of the time difference between the detection light and the pump light, the time delay system uses multiple reflective prisms to increase the propagation path of the detection light, thus adjusting the light path difference between the detection light and the pump light. It is noteworthy that the size of the optical path difference is usually closely related to the thickness of the tested sample, and by adjusting the optical path difference, it can effectively reduce or eliminate the time difference between the two. The resulting terahertz wave is received on the detector, obtaining the change of the terahertz pulse electric field intensity in the time axis, the terahertz signal will delay and the decay of the amplitude [20]. After the Fourier transform processing, the time domain waveform received by the detector can be converted into the frequency domain spectrum. By comparing the frequency domain spectral lines of the pump light and the detection light, we can obtain the relevant optical information, and obtain the refractive index, reflectivity and absorption coefficient that can represent the physical structural information and chemical properties of the tested sample through the extraction of optical parameters.

3. APPLICATION OF TERAHERTZ TECHNOLOGY IN FOOD SAFETY TESTING

At present, food safety has become one of the hot issues of concern, harmful residues in food quality and safety. Harmful residues mainly include foreign body residues, antibiotic residues, mycotoxin residues and pesticide residues.

3.1 Resi of Foreign Body

Ok et al. [21]used polygon and f-thetha lenses for food detection in the Asia-Pacific Hertz transmission imaging system and showed a fast scanning speed of 80 mm/s, a high resolution of 210 GHz and a detection range of about 150 mm. Shin et al. [22] used the complex refractive index mapping experiment in THz-TDS (0.2-1.3 THz) to identify insects as foreign bodies in various foods.It is impossible to classify insects by using the mapping concept because the refractive index and absorption coefficient of different insects are equivalent. However, insects can be detected from food in terahertz imaging. In addition, terahertz reflectance and imaging technology identified Tenebrio molitor in sugar rice and salt In the range of 0.3-1.2 THz, a new method based on dielectric traces is realized to distinguish food materials from foreign bodies such as insects Even the impurities in wheat, such as wheat husk, wheat straw, wheat leaves, wheat grains, weeds and ladybugs, were identified,Shen et al.[23] used terahertz frequency domain imaging combined with wheat V2 convolution neural network (CNN) model, and the recognition accuracy was 97.83% in the range of 0.2-1.6 THz .Hu et al.[24] used CARSUVE and SPA to extract the spectral features of foreign bodies in fish and optimized the model. The accuracy rate of prediction set reached 99.56% The THZ images of samples in the range of 0.1-4.0 THZ are visualized and the visualization of fish foreign bodies by THZ technology is realized .Ge et al.[25] used terahertz spectroscopy and deep support vector machine (DSVM) to detect five heavy metal pollutants, namely arsenic, lead, mercury, chromium and cadmium, in wheat samples .Compared with deep neural network (DNN) and SVM model, the comprehensive evaluation accuracy of DSVM model is 91.3%, which improves the classification accuracy of heavy metal pollutants in wheat .It is proved that terahertz technology is feasible in detecting foreign matter residues in food.

3.2 Antibiotic Residues

Antibiotics and their harmful residues have attracted much attention in recent years. Tetracycline hydrochloride (TCH) is an antibiotic widely used in the treatment and prevention of microbial infection in animals. Qin et al.[26] tried to use terahertz-TDS combined with PLSR to detect and quantify TCH in powder and solution samples in the range of 0.4 ~ 2.0 THz. Tetracycline hydrochloride (TC-HCl) is cheap, stable and easy to be absorbed and decomposed into tetracycline (TC) .The results show that the PLSR model of powder sample is very successful in controlling the TC, Rc = 0.9972, RMSEC = 1.31%,Rcv = 0.9919,RMSECV = 2.23%,RPD = 7.87. However, PLSR model is not robust to solution samples and needs to be improved in the future .Then Qin et al.[27] incorporated attenuated total reflection(ATR) technique into terahertz-TDS to determine the complex refractive index of tetracycline hydrochloride (TCH) solution.They tried to establish a model for predicting TCH concentration at 0.5 terahertz combined with SLR, and found that the established models had high R^2 values (0.95 ~ 0.98) and low RMSE values (0.61 ~ 0.99 mg/mL) and low detection limit (LOD) ranged from 0.45 ~ 1.29 mg/mL.The results show that ATR THz-TDS can realize liquid samples that THz-TDS is difficult to measure due to its high water absorption in terahertz region.In addition, for the strong localization and enhanced field of metamaterials, Qin et al.[28] proved that metamaterials composed of a group of annular pores deposited on metal films can be used as highly sensitive sensors for detecting trace TCH (as small as 0.1 mg/L) in aqueous solutions at terahertz. It is about 105 times higher than that determined on silicon material, which is equivalent to the maximum residue of TC antibiotic in food matrix Prove the potential of terahertz technology in detecting antibiotic residues in food .Bai et al.[29] used terahertz spectroscopy and imaging technology to quantitatively evaluateBackpropagation Neural Network (BPNN) to obtain the best imaging.The relationship between gray value and concentration of different mixtures under imaging was established .The binary mixture of fluoroquinolones (FQs) with different concentrations in fish meal feeds (FMF) has the best imaging effect and high accuracy at the characteristic absorption of 0.825 THz.Zhang et al.[30] designed a multi-band terahertz (THz) metamaterial sensor (TMS) with specific structure and proposed a multi-resonance response competitive and selective fusion (MRR-CSF) framework.The detection accuracy of enrofloxacin (ENR), pefloxacin (PEF) and nafloxacin (NAD) increased by 21% ~27%, 13% ~ 17% and 12% ~ 13%, respectively. The detection limit of ENRPEF and NAD antibiotic solution was 5 ng/L. The reliable recovery range was 100% ± 25%. The average recovery was more than 90% and the relative deviation was less than 5%.

3.3 Mycotoxin Residue

Fungal contamination is widespread, but its main concern is safety. Aflatoxin is a mycotoxin mainly produced by fungi, which is widely found in agricultural products and foods. Ge et al. [31] quantitatively determined the concentration of aflatoxin B1 in acetonitrile solution using linear (partial least squares (PLS) andprincipal component regression (PCR)) and nonlinear (nonlinear (support vector machine (SVM) and PCA-SVM) regression models using THz-TDS of 0.4 ~ 1.6 THz. The results showed that the linear regression model was superior to the nonlinear regression model in the range of aflatoxin B1 concentration from 1 to 50 μg/mL, and the results of the nonlinear regression model in the range of 1 to 50 μg/mL were more accurate, which laid the foundation for the application of THz technology in food mycotoxin detection. Liu et al.[32] used THz-TDS spectroscopy combined with BPNN and t-distribution random intercalation (t-SNE) of 0.1 ~ 2.5 THz to detect aflatoxin B1 exceeding 2μg/kg in soybean oil, with an accuracy rate of 90%, demonstrating the potential of THz technology in quantitative detection of food mycotoxins. In order to detect aflatoxin in food more accurately and quantitatively. Zhao et al.[33] used THz-TDS and metamaterial-based terahertz biosensors for quantitative determination of aflatoxins B1 and B2 in the range of 1.0 ~ 1.5 THz. The physical properties of aflatoxins B1 and B2, including refractive index, absorption coefficient, and dielectric function, were measured using Terahertz time-domain spectroscopy(THz-TDS) (transmittance) and Maxwell-Garnett useful medium theory. The content, thickness and dielectric constant of aflatoxin were measured. Mycotoxin B2 was found to have a higher frequency shift than aflatoxin B1, which helps to distinguish toxins. In addition, the volume of micro-upgraded toxins is identified and detected in this technology, and it also provides a new method for the detection of fungal content in food. Hu et al.[34] designed an "X" bimodal structure terahertz metamaterial sensor based on electromagnetic theory by using the essence of metamaterial structure enhancement, collected the terahertz transmission spectra of aflatoxin B2 solution with different concentrations, and used chemometrics to model the terahertz transmission spectra of aflatoxin B2 solution with full concentration. Highly sensitive, rapid and non-destructive detection of aflatoxin B2 solutions was achieved.

3.4 Pesticide Residues

In modern agriculture, pesticides are increasingly used to improve crop yields. However, pesticide residues can cause pollution and health problems.In a study, Ma et al.[35] used four variable selection algorithms of partial least squares(PLS), interval PLS(iPLS), backward iPLS(biPLS) and moving window PLS(mw PLS), to predict the content of thiabendazole based on THz-TDS spectrum, and found that the absorption spectrum of thiabendazole in the content range of 0% ~ 50.21% was completely distinguished, the relative error of mwPLS model in the range of 1.06 ~ 1.22 THz was 3.75%, the root mean square error of cross validation(RMSECV) was 0.2392, the root mean square error of prediction(RMSEP) was 0.3629, and the prediction accuracy was very high. In another study, Wang and Ma[36] used PLS, iPLS, biPLS, and mwPLS combined with THz-TDS spectroscopy at 0.3 ~ 1.6 THz to evaluate the content of nitrophene. Compared with the other 3 models, the biPLS model has the best prediction effect of RMSECV = 0.3157, RMSEP = 0.4064, correlation coefficient(R)= 0.9995 in the intervals of 0.907-0.943 THz, 0.962-0.998 THz, and 1.071 ~ 1.108 THz. However, PLS performs the worst, probably due to the inevitable noise brought in from the entire terahertz spectrum. In addition, Baek et al.[37] explored the feasibility of THz-TDS detection of methomyl in wheat, and selected the characteristic terahertz absorption peak at terahertz as the fingerprint for determining methomyl content in food matrix. The calibration curve for methomyl showed an The calibration curve of Dedudway shows Coefficient of Determination (Rc^2) > 0.957, but the sensitivity of this test for methomyl remained low with a Level of Detail (LOD)< 3.74%. Furthermore, Chen et al.[38] implemented a terahertz-tds system in the range of 0.3-1.7 THz to quantify imidacloprid in rice flour samples. AsLS method is used to correct the terahertz absorption coefficient spectral lines, and the signal-to-noise ratio of the spectral lines is improved.After that, PLS, SVM, iPLS and biPLS were used to establish quantitative models and predict them. All achieved satisfactory results, with an average RMSEP < 0.7% and a minimum LOD of 0.99%, indicating that this method can be used for the quantification of imidacloprid in rice, demonstrating the potential of THz technology in quantitative detection of pesticides. Recently, Lee et al.[39] reported a novel small molecule sensing tool for highly sensitive pesticide residue detection using THz-TDS of nanoslot antenna array, and found that even 8 ppm of methandomine in solution could be detected by enhanced terahertz spectroscopy. Furthermore, methomyl at a concentration of 1000 ppm remaining on real apple peel was successfully shown by 1.0 THz reflectance imaging. In terms of the penetration ability of terahertz waves, this technology also provides the possibility of real-time determination of pesticide residues in fruits, and its penetration depth can reach several millimeters. It can not only realize the rapid and nondestructive detection of pesticide content in fruits, but also its prediction accuracy is very high, which provides a new research for the detection of pesticide residues in food. Dai et al.[40] proposed a metal-graphene hybrid metamaterial THz sensor, and the designed metamaterial structure supports the excitation of ring dipoles under resonance. The detection limit used for drip-dried chlorothalonil solution was as low as 100 pg/mL.

4. CONCLUSION

With the rapid development of food economy, people have focused on studying the rapid and non-destructive detection methods of food safety testing. Terahertz spectroscopy technology has a unique advantage in the field of food safety. It can realize the high-resolution detection of food composition, structure and morphology, and performs well in the detection of major components such as water, fat and sugar. Terahertz spectroscopy also has the characteristics of fast, non-destructive and high sensitivity, which avoids the possible cross-contamination problems of traditional methods in the detection process, and improves the detection efficiency and accuracy. The unique advantages of THz make it stand out from both traditional and modern detection techniques. Although terahertz spectroscopy has better results, it has many limitations on water absorption, transmission depth, scattering defect, low detection limit, high signal to noise ratio, particle size and sample surface roughness compared with other traditional spectral detection. It is believed that the terahertz spectral technology will be further improved, which will play an increasingly important role in the field of food safety testing, and make more contributions to ensuring food safety and promoting the development of the food industry.

DISCLAIMER (ARTIFICIAL INTELLIGENCE)

Author(s) hereby declare that NO generative AI technologies such as Large Language Models (ChatGPT, COPILOT, etc) and text-to-image generators have been used during writing or editing of manuscripts.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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