Terahertz spectra of edible pigments

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Terahertz time-domain spectroscopy (THz-TDS) is a new coherent spectral technique to measure transmission spectra and analyze the spectral characteristics of samples. In this letter, the transmission spectra of edible pigments in the THz frequency range are measured using THz-TDS technology at both room temperature and low temperature in nitrogen atmosphere. Their refractive index spectra and absorbance are obtained. The result shows that there is some obviously abnormal dispersion in the refractive index spectra as well as absorption peaks in absorbance at the corresponding frequencies. THz-TDS is expected to have a potential application in the identification of pigments.

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Food security attracts extensive attention these days. It relates not only to personal health but also to the development of economics and society. As one of the most important food additives, edible pigments have a wide application in food coloration. Numerous methods are used to inspect the component of compounds, such as HPLC^[1], fast gas chromatography^[2], high performance liquid chromatography^[3], etc., but all these methods may cause the loss of sample functions. In this letter, we investigate the optical characteristics of some edible pigments at both room temperature and low temperature in nitrogen atmosphere experimentally by terahertz time-domain spectroscopy (THz-TDS) technique, which is carried out as a non-destructive spectral detection of materials.

The THz frequency range of an electromagnetic spectrum is typically identified as 0.1-10 THz^[4]. In the past decade, the experimental technique of THz-TDS has proven to be a versatile method for investigating a wide range of phenomena in the THz or far infrared spectral region and is always considered an effective method to determine a sample's refractive index and absorption coefficient at THz frequencies^[5-9].

Cochineal powder, whose main component is carminic acid (Formula: $C_{22}H_{20}O_{13}$), is a kind of pigment with extensive applications in the food coloring of wine, fruit juice concentrate, drink, sweet, sausage, etc. The samples used in this experiment were cochineal powder made from DINGHAO (DH) and KING. All the samples were pressed into several slices with a diameter of 13 mm and thickness less than 1 mm under the pressure of 4 tons after grinding. The surfaces of the slices were smooth and parallel.

THz spectra of the above samples were measured on a standard THz-TDS setup^[10,11]. Ambient temperature and humidity can affect the characteristics of a sample. It is mainly manifested in the variation of absorbance in three aspects: enhancement of absorption, appearance of new peaks, and blue shift of known peaks. To investigate these different properties of samples at low temperature, we improved the THz-TDS setup with a low temperature system. When the low temperature system works, the vacuum of the chamber decreases to about 10^{-5} Pa. The temperature can be minimized to 7.8 K or remain at a certain temperature.

After transmitting through the sample, the THz beam carries both amplitude and phase information. We define the signal detected without samples as the reference signal and that with samples as the sample signal. All the signals are transformed into the frequency domain by the fast fourier transform $(FFT)^{[12]}$. Therefore, the refractive index $n(\omega)$ can be obtained with the known thickness d and the ratio between the phase of sample and reference $\Phi(\omega)$ by

$$n(\omega) = \frac{\Phi(\omega)c}{\omega d} + 1, \qquad (1)$$

where ω is angular frequency, and c is the velocity of light^[13]. Thus, the extinction coefficient $\kappa(\omega)$ and absorption coefficient $\alpha(\omega)$ can be obtained as

$$\kappa(\omega) = \ln\left[\frac{4n(\omega)}{\rho(\omega)\left[n(\omega)+1\right]^2}\right]\frac{c}{\omega d},$$
(2)

$$\alpha\left(\omega\right) = \frac{2\kappa\left(\omega\right)\omega}{c} = \frac{2}{d}\ln\left[\frac{4n\left(\omega\right)}{\rho\left(\omega\right)\left[n\left(\omega\right)+1\right]^{2}}\right],\qquad(3)$$

where $\rho(\omega)$ is the ratio of amplitude between the sample signal and the reference signal^[14,15].

In the experiment, we first studied how the purity of edible pigments affected THz signal due to its strong absorption to the THz signal. Polyethylene (PE) is a material with high transmissivity to THz and is almost transparent in the low frequency of the THz frequency range^[16,17]. We mixed the pigment powder with the PE powder with weight ratios of 100%, 20%, and 10%, and pressed them into three slices with a thickness of 639, 762, and 786 μ m, respectively. As shown in Fig. 1, a stronger amplitude and a wider effective bandwidth of the spectra are obtained with decreased pigment purity.



Fig. 1. THz spectra of pigments in different proportions doped with PE.

The transmission spectra of DH and KING cochineal powder were measured by THz-TDS technology at both room temperature and low temperature in nitrogen atmosphere. The purity of the samples was maintained at 20%, and their thickness was 720 and 593 μ m, respectively. The refractive index of both samples was calculated by Eq. (1), and the results are shown in Figs. 2 and 3. Solid lines indicate the refractive index at low temperature, whereas the dashed lines denote that at room temperature. The averages of the refractive indexes of both samples are about 1.52 and 1.39, respectively. There are several abnormal dispersions in the spectra.

The absorption coefficient of the two samples was calculated according to Eqs. (2) and (3). Their absorbance



Fig. 2. Refractive index spectra of DH cochineal powder at different temperatures.



Fig. 3. Refractive index spectra of KING cochineal powder at different temperatures.



Fig. 4. Absorbance of DH cochineal powder at different temperatures.



Fig. 5. Absorbance of KING cochineal powder at different temperatures.

is presented in Figs. 4 and 5, respectively. Clearly, the absorption peaks correspond to the abnormal dispersion in Figs. 2 and 3.

In the above experiments, we obtain the refractive index spectra and absorbance of DH and KING cochineal powder, respectively. For DH cochineal powder, the refractive index is about 1.51 at room temperature and 1.52 at low temperature. Two curves in Fig. 2 have the same variation trend. In Fig. 4, there are two peaks at room temperature located at 1.24 and 1.58 THz, and three peaks at low temperature located at 1.25, 1.58, and 2.40 THz, respectively. The peak position at 1.58 THz does not change against the temperature. However, a tiny blue shift occurs in the peak position at 1.24 THz at room temperature and 1.25 THz at low temperature. At a frequency of 2.40 THz, a new peak appears at low temperature. This is due to the spectral broadening at high temperature while the absorption peaks at low temperature sharpens so that they can be recognized. Comparing the absorbance of the two samples, there is a similar absorption peak located at 1.25 THz, which is probably due to the absorption of their same component, carminic acid. Other absorption peaks are caused by the different impurities from different manufacturers.

Similarly, the refractive index of KING cochineal powder is about 1.38 at room temperature and 1.40 at low temperature; an absorption peak appears at low temperature located at 1.25 THz. Therefore, THz spectra at low temperature can recognize spectral properties of materials in more detail.

In summary, THz spectra of some edible pigments are

measured by THz-TDS at room temperature and low temperature, respectively. The results show that the pigments have certain fingerprint absorption and refractive index spectra in the THz region. THz-TDS can be applied to obtain information on the absorption characteristics of samples. Therefore, it provides a new method of non-destructive inspection of edible pigments and assists in the identification of pigments.

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