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Terahertz Dielectric Properties of Polymers

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The terahertz dielectric properties of polymers were characterized by transmission terahertz time-domain spectroscopy (THz-TDS) in the frequency range extending from 0.2 to 3.0 THz. The terahertz absorption spectra, the refractive indices and the dielectric functions of various polymer materials were measured and compared. The variation of the refractive index of the polymers was less than 6 %, ranging from 1.4 to 1.8, within the investigated frequency range, but the absorption properties of the polymers showed very different frequency-dependent behaviors. The loss mechanism for terahertz radiation in polymers is discussed by correlating the absorption coefficients and the loss tangents ($\tan\delta$) of the materials.

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I. INTRODUCTION

Over the past ten years, there has been a remarkable effort in employing terahertz (THz) spectroscopy for investigating material properties [1–4]. Pulsed THz time-domain spectroscopy (THz-TDS) is a coherent technique, in which both the amplitude and the phase of a THz pulse are measured. Coherent detection enables direct calculations of both the imaginary and the real parts of the refractive index without using the Kramers-Kronig relations. From the resulting refractive index, one can obtain the absorption coefficient and the dielectric constant of the material. THz-TDS has been employed to investigate a wide variety of materials, including liquids [5], semiconductors [6,7], superconductors [8], explosive materials [9], and gases [10].

Various polymers have been widely used for packages, bottles, semiconductor devices, electrical insulation, and other applications. The electrical properties below a few gigahertz and the optical properties of polymer materials are well known, but there are few reported data on the properties in the far infrared or terahertz ranges. The refractive index and the absorption coefficient for polyethylene and teflon at the millimeter wavelengths are discussed in Ref. 11. Recently, Nagai *et al.* reported the dielectric properties of polyamide (PA-6) in the THz frequency region [12]. In this article, the THz optical properties and the complex dielectric function of several polymer materials are experimentally characterized by using transmission THz-TDS. The refractive index, the power absorption, and the complex dielectric functions in the THz region are compared with known values for each material. From the measured dielectric properties, the loss mechanism of THz radiation for each polymer is also discussed.

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II. EXPERIMENTS

1. THz Time-Domain Spectroscopy System.

A standard transmission THz TDS system is employed and is illustrated schematically in Fig. 1 [13]. The laser source is a commercial mode-locked Ti:sapphire laser (Spectra Physics Mai Tai) producing 70 – 120 fs pulses at 780 – 920 nm with a repetition rate of 80 MHz and an average power of 600 – 1200 mW. The fs laser beam was divided into two beams, one for exciting the emitter antenna, and the other for measuring the THz signal at

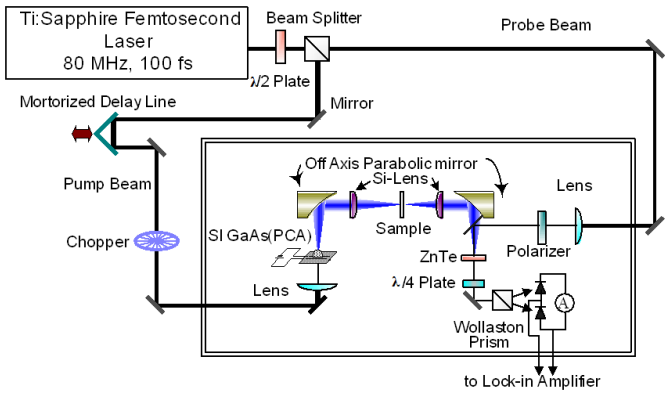

Fig. 1. Experimental setup for transmission THz time-domain spectroscopy.

Table 1. List of polymer samples and their physical and electrical constants [16].

Abbreviations	Full Name	Refractive Index	Dielectric Constant @1 MHz
PP	Polypropylene	1.49	2.2 – 2.6
PTFE (Teflon)	Poly tetrafluoroethylene	1.38	2.0 – 2.1
HDPE	Polyethylene-high density	1.54	2.3 – 2.4
ABS	Polyacrylonitrile - butadiens - styrene	NA	3.2 – 3.4
PET (Polyester, Mylar)	Polyethylene terephthalate	1.58 – 1.64	3.0
Polyaramid	Polymetaphenylene isophthallamide	NA	4.4@60 Hz
PMMA, Acrylic	Polymethlmethacrylate	1.49	2.6
PC	Poly carbonate	1.584 – 6	2.9
PA 6	Polyamide nylon 6	1.53	3.6

the detector crystal. The THz emitter is a coplanar strip line antenna fabricated on a semi-insulating (SI) GaAs wafer. The gap distance of the emitter antenna is 200 μm and is excited with a 7-mW of focused laser beam. The bias voltage between the coplanar strip lines of the emitter antenna is 30 V (E_{bias} = 1.5 kV/cm). An aplanatic hyper-hemispherical silicon lens is attached to the back of the THz emitter to increase the coupling efficiency of THz pulse from the antenna to free space. Two highly resistive silicon lenses are used for focusing the THz beam to samples with a spot size of less than 2 mm diameter. The detector crystal is 0.5-mm thick <110> ZnTe and is gated by 1-mW probe beam pulses. A standard electro-optic sampling (EOS) setup is used for THz signal acquisition [14]. To increase the signal-to-noise ratio, we modulate the pump beam with a mechanical chopper at 2 kHz. In order to prevent absorption by atmospheric water vapor, we enclose the THz generation and detection components in an acryl box supplied with dry air. The output signal from the balanced diode detector is monitored with a lock-in amplifier and a computer.

2. Calculations of The Complex Refractive Index and The Dielectric Function

In THz-TDS, we are interested in measuring $\tilde{\varepsilon}(\omega) = \varepsilon^{'}(\omega) - j\cdot\varepsilon^{''}(\omega)$ or equivalently $\tilde{n}(\omega) = n(\omega) - j\cdot\kappa(\omega)$which is the complex index of refraction of the material, where $\tilde{\varepsilon}(\omega) = (\tilde{n}(\omega))^2$. The complex frequency spectra of the reference, $\tilde{S}_r(\omega)$, and of the sample, $\tilde{S}_s(\omega)$, are calculated using numerical Fourier transforms from the time-domain waveforms sampled in the experiment. The analytic expressions for n_s and κ_s can be written in terms of the magnitude $\rho(\omega)$ and the phase $\phi(\omega)$ of the ratio of $\tilde{S}_s(\omega)$ to $\tilde{S}_r(\omega)$ as follows [15]:

$$\frac{\tilde{S}_s(\omega)}{\tilde{S}_r(\omega)} = \rho(\omega)\cdot e^{-j\phi(\omega)} \tag{1}$$

$$n_s(\omega) = \phi(\omega)\cdot\frac{c_0}{\omega d} + 1 \tag{2}$$

$$\kappa_s(\omega) = \ln\left(\frac{4n_s(\omega)}{\rho(\omega)\cdot(n_s(\omega)+1)^2}\right)\cdot\frac{c_0}{\omega d} \tag{3}$$

Here, d (cm) is thickness of the sample, and c_0 is the speed of light in vacuum. The imaginary part of the refractive index, κ_s, is related to the power absorption coefficient, $\alpha(\mathrm{cm}^{-1})$, as

$$\kappa_s = c_0\alpha(\omega)/2\omega$$

where

$$\alpha_s(\omega) = \frac{2}{d}\cdot\ln\left(\frac{4n_s(\omega)}{\rho(\omega)\cdot(n_s(\omega)+1)^2}\right) \tag{4}$$

Hence, the real and the imaginary parts of the dielectric constant are determined from the following equations:

$$\varepsilon_s^{'}(\omega) = (n_s(\omega))^2 - [c_0\alpha(\omega)/2\omega]^2 \tag{5}$$

$$\varepsilon_s^{''}(\omega) = 2n_s(\omega)\kappa_s(\omega) = c_0 n_s(\omega)\alpha_s(\omega)/\omega \tag{6}$$

III. RESULTS AND DISCUSSION

In the present study, nine kinds of polymer materials were examined. All specimens were manufactured by Goodfellow Co. and are listed in Table 1. The physical

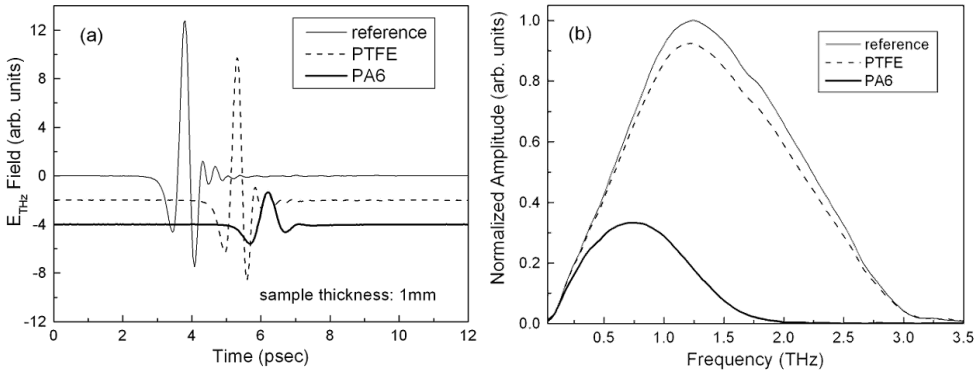

Fig. 2. (a) Measured THz pulses from the reference and the sample. For clarity, the sample pulses are vertically displaced. (b) Corresponding amplitude spectra of the measured reference pulse and the sample pulses.

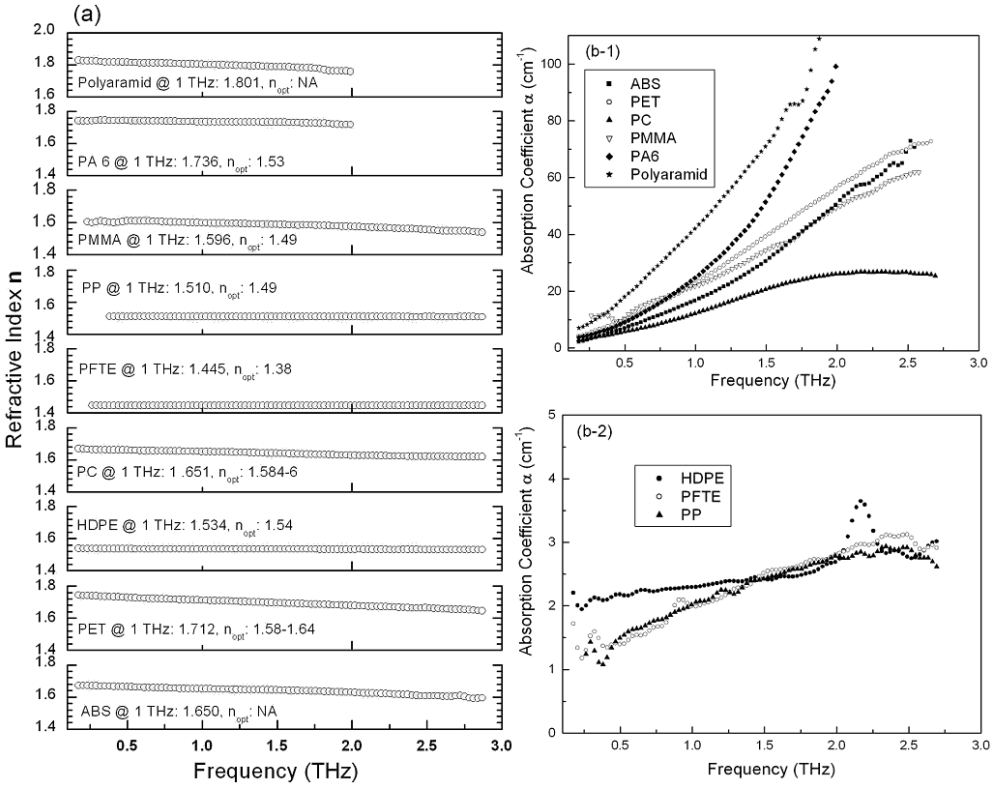

Fig. 3. (a) Refractive index and (b) absorption coefficient of the polymer materials.

and the electrical constants in Table 1 were provided by the manufacturer [16].

The transmitted electric field of terahertz pulses through the sample and the reference are recorded in the time domain, and the corresponding frequency spectra are obtained by using numerical Fourier transforms. Fig. 2 shows (a) the measured reference and sample (PTFE, PA6) pulses and (b) the corresponding Fourier transformed spectra. Every pulse curve is the average of three individual measurements in order to increase the signal-to-noise ratio. There is no absorption peak originating from water molecule in the reference spectrum [17], so we can ignore the water absorption effect. The signal-to-noise ratio in the measurement is estimated to be higher than 1000.

The refractive indices $n_s(\omega)$ and the power absorption coefficients $\alpha_s(\omega)$ of the nine polymer samples are calculated from the measured data by using Eqs. (2) and (4). The calculation results are plotted in Fig. 3. The absorption curves are grouped according to type for easier comparison. The refractive indices of the nine samples show slow a decrease, but the variation is less than 6 % within the investigated frequency range. In Fig. 3 (a), the refractive indices of the materials at 1 THz and at

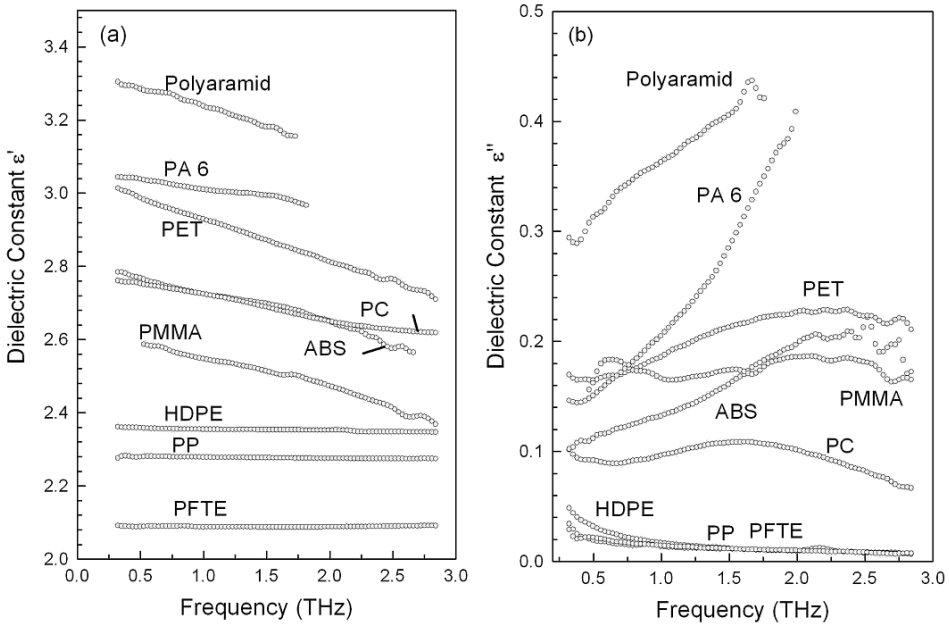

Fig. 4. (a) Real and (b) imaginary parts of the dielectric constant

the optical frequency (given by Table 1) are compared. All the refractive indices at 1 THz are a little higher than those at the optical frequency, except for HDPE, and we can expect the refractive index in the terahertz range to approach the optical value at higher frequencies.

Meanwhile, the power absorption curves show very different behaviors for different materials. The absorption of polyaramid, ABS, and PA 6 increase quadratically with frequency, but those of HDPE, PTFE (Teflon) and PP are very weak and nearly constant in the THz range. We suppose that the high absorptions of the above polymers are related with the high dielectric loss, which will be explained below. The slowly varying refractive indices and the small absorption coefficients of HDPE, PTFE and PP in the terahertz region enable them to be good candidates for THz optical materials such as lens and windows [11].

The real and the imaginary parts of the dielectric constant can be calculated from Eqs. (5) and (6). Fig. 4 shows the recorded data for the complex dielectric constants of the nine samples. In Fig. 4 (a), the real dielectric constants of HDPE, PTFE, and PP show a feature that is essentially the square of the dielectric constant $n_s(\omega)$ because the absorptions by the samples are small in the spectral region concerned and the contribution of the absorption coefficient to the real dielectric constant is nearly negligible in Eq. (5). Furthermore, we can see that the real dielectric constant at low frequency obtained by a linear fit of the measured data is in good agreement with the given value at 1 MHz in Table 1.

The power loss in the dielectric can be expressed in terms of the loss tangent ($\tan\delta = \varepsilon^{''}/\varepsilon^{'}$). Fig. 5 shows the relation between the power absorption coefficient and $\tan\delta$ at 1 THz. A strong correlation between the absorption coefficient and the loss tangent was observed, and this result indicates that the main mechanism of THz field absorption is the dielectric loss caused by molecular collisions and vibrations. The consistency of the measured data for a the given material property suggests that our THz-TDS measurement is exact. As a result, the refractive index and the dielectric constant in the THz range acquire a new meaning.

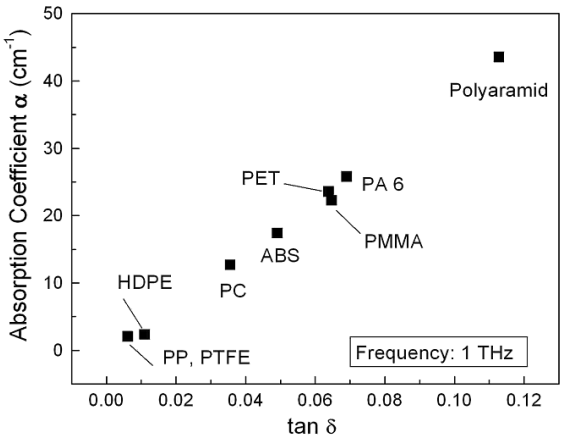

Fig. 5. Relation between the absorption coefficient and the loss tangent ($\tan\delta$).

IV. CONCLUSIONS

The dielectric properties of various polymer materials in the THz region have been investigated systematically

by using a transmission THz-TDS method. The refractive indices, the power absorption coefficients, and the complex dielectric constants of the polymers were measured and analyzed. New optical and electrical data for the polymers were obtained, and the consistency with the known parameters was identified. The results obtained in this study suggest that the THz-TDS method is a useful tool for polymer characterization in the far infrared region.

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