Infrared Physics & Technology 89 (2018) 110–114

Contents lists available at ScienceDirect

Infrared Physics & Technology

journal homepage: www.elsevier.com/locate/infrared

A terahertz transmission imaging based approach for liquid alcohol wettability investigation

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HIGHLIGHTS

- Obtain liquid depth distribution from terahertz transmission image content.
- Our method can compare material's surface wettability besides the contact angle.
- The technique can be used for biomedical field with minimal health risk.

ARTICLE INFO

Article history:
Received 17 July 2017
Revised 30 December 2017
Accepted 30 December 2017
Available online 2 January 2018

Keywords:
Terahertz imaging
Image noise reduction
Polynomial fitting
Wettability

ABSTRACT

By using terahertz time-domain spectroscopy (THz-TDS), the transmission spectral images of a series of aliphatic monohydric liquid alcohols were investigated in this work. The image gray value indicated the non-uniform distribution of the liquid. After noise reduction, the depth distribution was computed from the image content, which depended on the wettability of the container surface by the liquid. Then the volume of the liquid crept up to the container surface was compared quantitatively. Our result showed that the surface wettability of the PS container surface by the liquids increased with the alcohol alkyl chain increasing, which was verified by the contact angle of the liquid and the surface. The study indicated that the THz image might offer a potential technique for detecting the wettability of liquid directly without an additional contact angle experiment.

1. Introduction

In recent years, there has been a tremendous interest in the field of basic and applied THz-frequency radiation research, because its applications covered biomedical diagnostics, material recognition and security control, etc. [1]. A sizable fraction of this effort has been focused on the exploitation of the fact that most chemical molecules in solid state have rich and distinct absorption spectra in the THz region [2]. Unlike many solid chemical compounds that possess characteristic THz fingerprint spectra, liquid samples always show broad and featureless spectra in THz range, making it relative difficult for identification and analysis [3,4]. Although the lack of distinct spectral features hindered specific identification of a liquid, efforts have been done to get some of its characters. Kindt et al. reported the frequency-dependent optical constants of highly absorbing polar liquids including water, methanol, ethanol, and 1-propanol utilizing femtosecond (fs) THz pulses [5]. The dielectric properties of series monohydric alcohols in THz region were investigated by Yomogida et al. [6,7]. Jepsen et al. demonstrated that it was possible to determine the alcohol content of water-ethanol mixture inside polyethylene terephthalate and glass bottles by THz reflection spectroscopy [8]. Recently, metamaterials and some engineered structures were suggested as THz sensor for monitoring the refractive index of the surrounding liquids [9,10]. On the other side, THz imaging technique has attracted considerable attention since it was firstly demonstrated by Hu and Nuss [11] because it has many practical applications in diverse areas, such as packaging inspection, quality control of materials, chemical composition analysis, and biomedical diagnostics [12–16]. The THz absorption image carries many information of the object, providing both the absorption and the spatial distribution information of the sample. This information was proposed to be used to analyze the layer thickness and spatial pattern of

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https://doi.org/10.1016/j.infrared.2017.12.021
1350-4495/

solid chemical component nondestructively [14,15]. Akimune et al. proposed and developed a method of THz chemical microscopy based on a sensing plate to visualize different ions in water solutions [17]. With the development of THz technique, fast THz array imaging is expected to be achieved in the close future [18] and bring a wider application of THz imaging.

In this paper, we propose to analyze the THz transmission spectral image of liquids, trying to find out some liquid properties from the image, such as surface wettability, which describes its adhesion on the container surface. Since alcohols are important organic compounds and usual solvents used in chemistry and biology, in our work, a series of liquid monohydric alcohols in cell culture cluster units (made from polystyrene (PS)) were taken as study model. THz transmission spectral image of the alcohols were measured by THz-TDS. The image gray value indicated the non-uniform distribution of the liquid. The depth distribution was computed from the image content, which depended on the wettability of the container surface by the liquid. Then the volume of the liquid crept up to the container surface was compared quantitatively. Our result showed that the surface wettability of the PS container surface by the liquids increased with the alcohol alkyl chain increasing. To verify the conclusion, the contact angles between the liquids and the surface were measured by an Attension Theta System. The results conformed with Shull's [19]. The study in this paper indicates that we can visually "read" the liquid wettability from the THz image, especially when it is not convenient to measure the contact angle. This would have potential applications in sample label-free detection or classification of multiple samples.

2. Methodology

As we know, the liquid depth in the container is not everywhere the same because of the adhesion of the alcohols liquid on the PS container wall. The liquid depth distribution in the container reflects the adhesion strength, which is related to the surface wettability. According to the Beer-Lambert law, the spectra strength of the THz pulse transmitted through the liquid is proportional to the path depths. Consequently, the depth of the liquid in a container can be computed through the THz transmission spectra strength which can be obtained by THz transmission spectra imaging system. After applying image noise reduction on the image, THz transmission spectra strength distribution in a 2-D field can be achieved. Afterwards, the liquid depth distribution on which the surface wettability will be analyzed is computed. To verify the above result, we use the gold standard method – surface contact angle – to quantify the liquid wettability, which is proposed by Thomas Young [20].

The consistency of the above two results will verify the feasibility of method that using the THz transmission spectra image to analyze the liquid wettability. In our work, the monohydric alcohols methanol (>99.5%), ethanol (>99.7%), 1-propanol (>99%) and 1-butanol (>99.5%) were used for the experiment (purchased from China National Medicines Corporation Ltd.). The following parts describe the experimental design.

2.1. THz transmission spectra imaging

The measurement was carried out on a fiber-coupled THz-TDS system (FiCO) (Z-Omega Terahertz Corp, USA) [21], which was capable to perform spectroscopy and imaging. The system was pumped by an external 1560 nm pulsed laser source with an average power of 380 mW and pulse duration around 90 fs, repetition frequency at 80 MHz. The laser source was coupled with the base unit. The THz emitter consists of a photoconductive dipole antenna and a silicon lens was used to collimate the THz beam out of the module. The receiver used silicon lenses to focus the incoming radiation on the electro-optic crystal to detect THz waves. Transmitter and receiver were independent and fiber coupled with the base unit. The considered frequency range in our experiment was from 0.1 THz to 1.0 THz. The FiCO system has been equipped with an automatic x-y positioning module, which enables the system to scan an area with the smallest spatial offset along both x and y directions, equal to 0.125 mm.

Quartz liquid cells with different thickness (400 μm and 600 μm) were used for THz absorption measurement. And Costar 96-well culture cluster (purchased from Corning Incorporate) was used as liquid container for THz imaging. The wells are flat bottom with lid to avoid the volatile of liquid and samples cross-contamination. The container is manufactured from optically clear virgin PS which shows good transparent to THz waves. The alcohols used in the measurements are nonslovents for PS [19]. The monohydric alcohols were put into four of a 96-well culture plate simultaneously (as shown in Fig. 1), with equal amount of 40 μl in each well using a pipette. The diameter of each well is 0.64 cm. The plate is made of PS which has negligible THz absorbance, whence the transmitted signal through PS without sample is taken as reference.

Before forming the THz image, we firstly obtained the absorption coefficient of the four alcohols with Beer-Lambert law [22]. We used the method described by Kindt [5], spectra of the alcohol liquids at room temperature were measured with use of the quartz cell placed in the THz beam path. The transmission of the THz pulse through the liquid in the sample cell was measured for different path depths. A fast Fourier transform was applied to the time domain data, after subtracting a small linear base line term and zero-padding to obtain 2048 points, which then yielded the power $P(d, \nu)$ of each frequency component ν of the transmitted pulse for each path depth d. The absorption coefficient α at each frequency was determined by [5]:

$$\ln P(d,\nu) = \ln P_0(d,\nu) - \alpha(\nu)d \tag{1}$$

In the THz imaging stage, the control software of the FiCO system visualized in the imaging window a hologram, in terms of the peak-to-peak amplitude of the measured signal, wherein the x-y coordinates denoting the corresponding sample position. In our work, we selected the image slice at the frequency of 0.9 THz.

2.2. Contact angle measurement

The contact angle (CA) values of PS were measured on an Attension Theta System (KSV Instruments Ltd., Finland). A 4 μl drop from a needle tip was dripped onto the PS surface. A digital camera

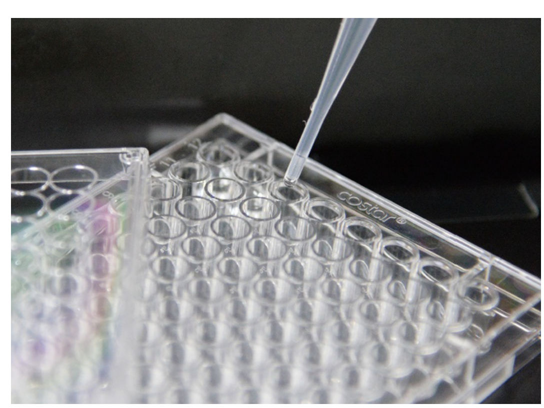

Fig. 1. The culture cluster used in our experiment and four cells contains the different alcohols.