Diagnosing water content in paper by terahertz radiation

D. Banerjee,[1] **W. von Spiegel,**[2*] **M. D. Thomson,**[2] **S. Schabel,**[1]
H. G. Roskos[2]

[1] *Chair of Paper Technology and Mechanical Process Engineering (PMV), Technische Universität Darmstadt, Alexanderstr. 8, D-64283, Darmstadt, Germany*

[2] *Physikalisches Institut, Johann Wolfgang Goethe-Universität Frankfurt, Max-von-Laue-Str. 1, D-60438 Frankfurt am Main, Germany*

w.v.spiegel@physik.uni-frankfurt.de

Abstract: We explore the application of terahertz spectroscopic techniques for the remote determination of the water content of paper. The aim is the development of a rapid diagnostic imaging tool applicable in paper fabrication processes. THz radiation offers a high sensitivity for water, a good spatial resolution, and insensitivity to scattering at the paper surface. The advent of THz cameras makes fast large-area image detectors feasible. In this paper, we show for the case of a 0.6-THz fixed-frequency system, that the water content of paper can be determined with high accuracy. We demonstrate a quantitative (calibrated) method for determining the moisture content in paper based on extinction and phase measurements in the lower THz range with a spatial resolution in the mm-range and scanning times below two minutes.

OCIS codes: (120.0120) Instrumentation, measurement, and metrology; (110.6795) Terahertz imaging; (120.7000) Transmission; (300.6495) Spectroscopy, terahertz

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1. Introduction

The drying process is a very important part of paper production because it has a strong impact on the final paper quality. The movement and distribution of the moisture during the drying process influences paper properties such as shrinkage, curl, strength, etc. Non-uniform moisture profiles can lead to various problems such as an increased fracture probability, difficulties upon reel formation, calendar blackening, printer misfeeding, and curling and cockling of paper. Online monitoring of the moisture content is hence an important task during the drying process.

With the increasing process speed and stringent quality requirements in modern paper production, a more detailed and accurate inspection of paper properties has become imperative. Especially, online information on the moisture distribution is needed as problems with moisture non-uniformity are known to occur with the prevailing drying technologies [1]. Existing monitoring techniques yield either spatially integrated or single-point data. The alternative solution of over drying the sheet during fabrication to avoid moisture non-uniformity is expensive because the drying curve becomes very steep towards the end of drying. It is preferable to directly produce sheets with a pre-defined moisture content and uniform distribution of the moisture.

Presently, common moisture meters are based on either microwave or near-infrared (NIR) devices. Microwaves exhibit good penetration and low scattering. The poor spatial resolution (due to the large wavelength) and inadequate accuracy at low moisture content are serious disadvantages [2]. The most popular techniques employ differential absorption measurements with multiple near-infrared frequencies [3]. The disadvantages are significant scattering, a poor penetration depth especially for heavier-stock papers, and the need for calibration because of the non-linear response as a function of moisture content and paper composition and grade.

In this paper, we describe spatially resolved moisture content measurements with a single-pixel scanning THz imaging system. With the advent of multi-pixel THz detectors [18, 19], it is principally possible to perform imaging of this kind in real-time. THz photonic technology currently provides imaging systems working at several THz [20, 21], where the extremely high absorption by bulk water may limit the application of THz imaging to very thin sheets of paper. On the low-frequency side of the THz regime, a number of companies offer real-time imagers which work at 0.1 THz, and at least one commercial system operates at several hundred GHz. None of these systems is built to measure amplitude and phase simultaneously, though. In the

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intermediate regime from about 0.4 THz to 2.5 THz, real-time THz imaging systems are not available, yet, but are currently being developed.

2. Measuring moisture content with THz

Compared to the common techniques mentioned in the introduction, there is a clear potential for improvement. Nearly a quarter century ago, far-infrared (sub-millimeter-wave) radiation was already explored as an interesting alternative, and a good accuracy of moisture content determination was found for light- and heavy-stock paper [4].

Far-infrared radiation, nowadays called terahertz (THz) radiation, is only weakly absorbed by paper, but strongly so by water [5, 6]. The spectral properties of water under various conditions are well known in the THz frequency range [7, 8, 9, 10].

The penetration depth of THz radiation in water is such that one obtains a strong absorption signal with moist objects with a thickness in the order of 100 μm (such as plant leafs) [11] which is ideally suited for measurements on paper. THz imaging can give sub-millimeter spatial resolution and is only weakly affected by scattering at material or surface irregularities which are much smaller than the wavelength [12, 13, 14]. The microscopic fibrous structure of paper hence is not resolved by THz radiation but seen as a homogeneous medium, and it does not lead to significant scattering.

In the following, we report on THz transmission measurements through moist paper performed with two types of measurement systems. The first is a time-domain spectroscopy (TDS) system based on a femtosecond Ti:sapphire amplifier laser with a pulse repetition rate of 1 kHz [15, 16]. It covers the frequency range of 0.1-2.5 THz. The second is a fixed-frequency THz imaging system based on a microelectronic radiation source, a Gunn emitter followed by a multiplier chain. It delivers radiation at 0.6 THz with a power of 0.5 mW. The electrooptic detection works with a mode-locked Ti:sapphire laser with a repetition rate of 82 MHz [17]. Imaging is performed in raster-scan mode: The sample is positioned on a translation stage and moved through the focus of the THz beam. Both systems allow us to measure the amplitude and phase of the THz radiation.

The paper samples are laboratory-made hand sheets prepared from spruce sulphite pulp of 80 g/m^2 (70 g/m^2 for TDS) with a thickness of 100 μm (90 μm for TDS). Prior to the measurements, the samples are impregnated in distilled water and then mounted on a vertical frame. Time series of THz transmission measurements are carried out as the paper dries in air. Between scans in the imaging setup, the actual moisture content of the sheet is determined gravimetrically, i.e. weighing of the sample with a precision (0.1 mg resolution) scale and comparing the actual weight with that of the sample after drying in an oven.

Figure 1 displays a time series of relative optical-density spectra measured with broadband THz pulses at a single spot of a moist sample during drying. Exemplary THz transients are shown in the inset. As the sheet dries over a time scale of one hour, the amplitude of the THz waveform increases. This is consistent with a reduction of absorption and reflection losses while the water content in the paper gradually decreases. The decrease of the index of refraction with the loss of water also leads to a temporal shift of the waveforms. The THz pulse arrives earlier at the detector when the water content is reduced. Hence, both the amplitude change and the phase change of the THz pulses can principally provide information on the water content of the sample.

The relative optical-density ($log_{10} \frac{P}{P_{ref}}$, where P is the transmitted power at the specified frequency, P_{ref} is the equivalent from the reference measurement) spectra are calculated from the Fourier transforms of the THz transients normalized to the Fourier data of the trace taken after 66 minutes neglecting changes in reflection. Data above 1.6 THz is not displayed because of the non-negligible influence of THz absorption by the water vapor of air in this range. The

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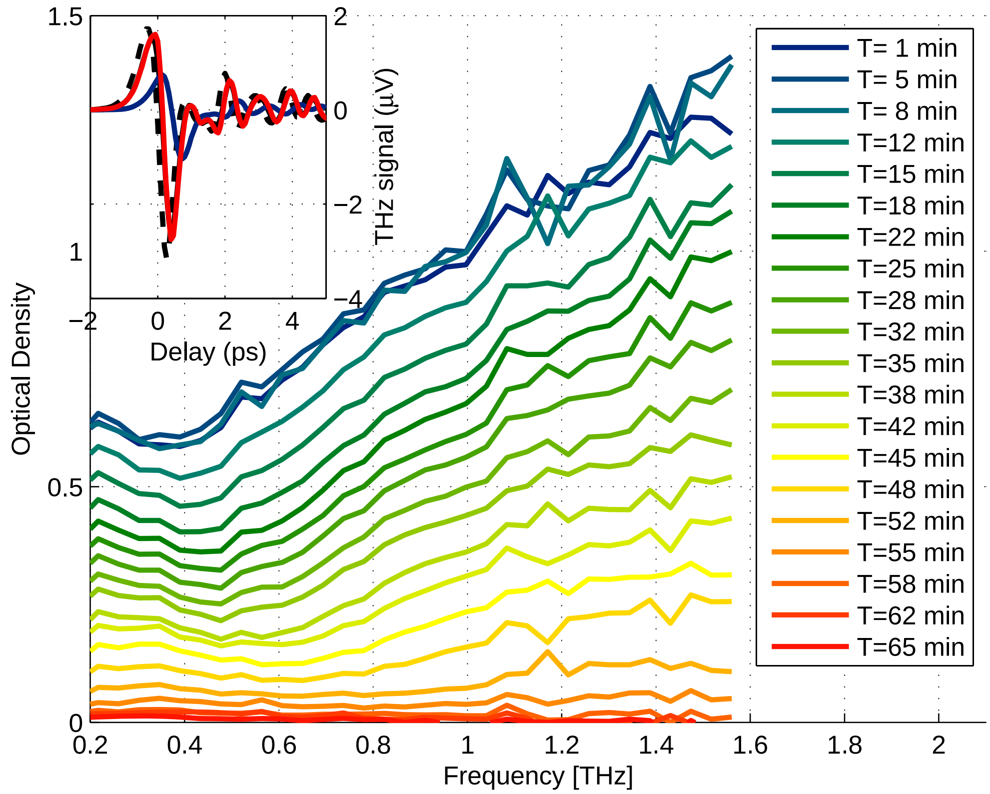

Fig. 1. Evolution of the frequency-dependent relative optical-density (relative to the measurement after 66 minutes) of a moist paper sheet drying. The spectra are calculated from THz transmission transients such as those shown in the inset. For the evaluation of the spectra, the full scan range of the transients of 20 ps is taken into account and not only the time window of 7 ps presented in the inset.

dip in the spectrum at 0.4 THz in the scan taken after 2 min shifting to 0.7 THz in the scan taken after 62 min probably represents a transmission maximum because of interference in the paper sheet. These effects are not examined in detail at the moment, but probably they can be taken into account to enhance the precision of moisture content measurements with this method.

The lower panel of Fig. 2 shows both the relative optical-density at 0.6 THz and the moisture content (specified in absolute values of kg water per kg fiber, from a typical drying curve gravimetrically determined with a different sample drying) as a function of drying time. A good correlation between the data is found. The upper panel displays the phase difference (between the transmitted amplitude at the specified frequency, taken from the Fourier spectra of the TDS data, and the equivalent from the reference measurement) of the THz signal relative to the measurement taken after 66 minutes. Again, a good correlation between phase change and the drying curve is observed.

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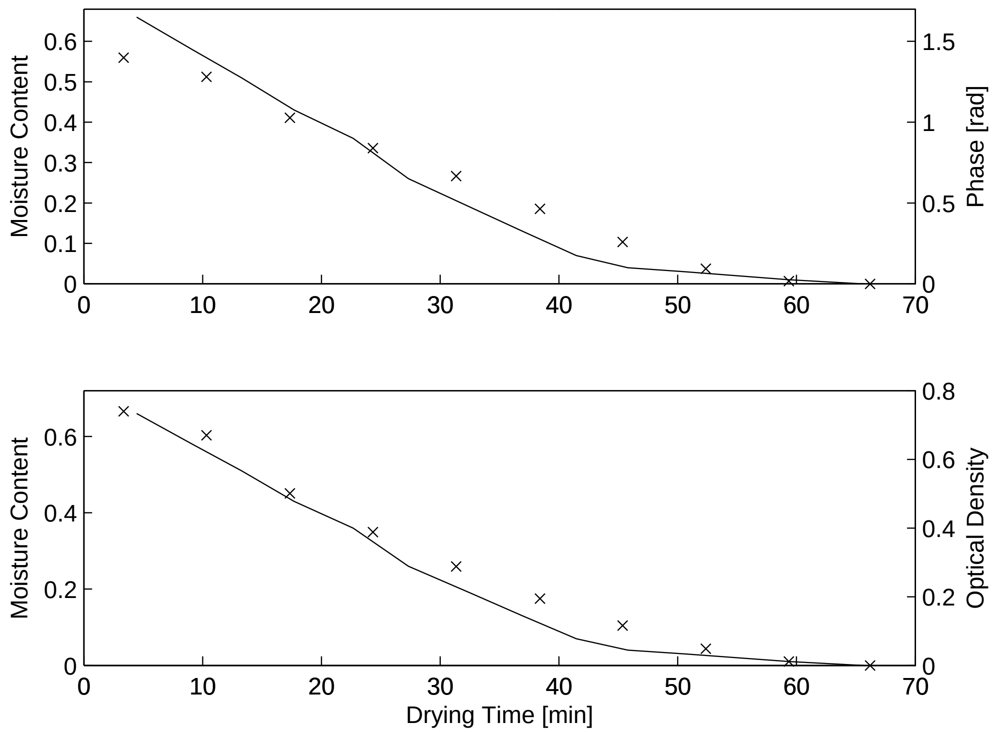

Fig. 2. Temporal evolution of the relative optical-density at 0.6 THz (×, lower panel) and of the phase change (×, upper panel, both from the TDS measurements, relative to the measurement after 66 minutes) compared to a typical drying curve of a different sample(—, gravimetrically determined with a precision scale).

3. Spatially resolved measurements

Imaging measurements are performed with the fixed-frequency setup. The 0.6-THz beam is focused onto the sample with a focal spot size of 1.2 mm. A dry and a water-impregnated paper specimen are mounted next to each other, and are scanned simultaneously (i.e. each line scan extends over both specimen). We repeatedly take images over an area of 40 mm × 40 mm which covers parts of both the dry and moist specimen. Data acquisition takes 1.7 minutes per frame (1600 pixels on a 1 mm grid).

The lower panel of Fig. 3 displays the relative optical-density ($-\log_{10}\frac{P_{cw}}{P_{cw,ref}}$, where P_{cw} is the transmitted power, calculated from the X and Y outputs of the lock-in amplifier measuring the intermediate-frequency signal, $P_{cw,ref}$ is the equivalent from the reference measurement) versus the gravimetrically determined water content, averaged over an area of $15 \times 40\,\mathrm{mm}^2$ on the moist sample. A linear function fits the data very well (coefficient of determination $R^2 > 0.99$). The same holds for the phase change (difference between the phase from the actual measurement and the phase from the reference measurement, both calculated from X and Y of the phase-sensitive intermediate-frequency measurement) as a function of water content (see upper panel of Fig. 3). Note that THz and gravimetric measurements are performed with the same sample alternately during the same drying process. The remarkable correlation between the data sets hence can serve for calibration of the THz-based moisture content measurements.

With this good quantifiability of water content, one can map the spatial moisture distribution

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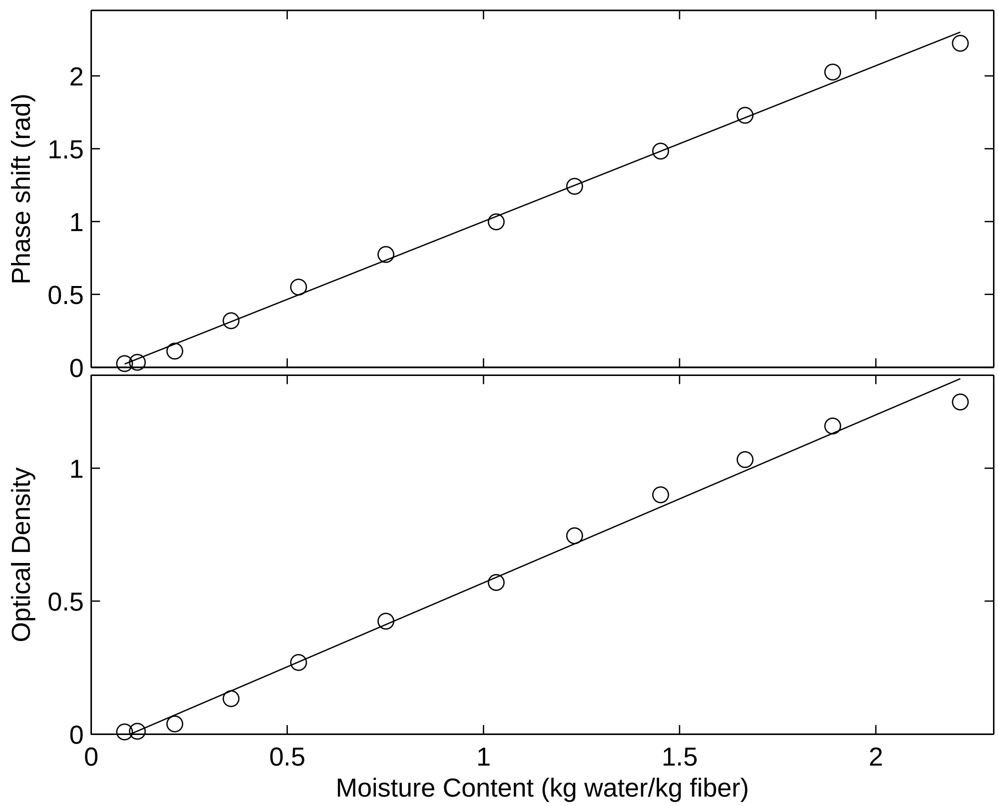

Fig. 3. Phase change (○, upper panel) and relative optical-density (○, lower panel) relative to the dry paper sample shown as a function of the water content in the moist sample. The linear regressions (—) linking the water content to the THz relative optical-density and phase shift, respectively, are also shown in the graphs.

in paper in absolute terms. Fig. 4 presents an image of a $80\,\mathrm{g/m^2}$ paper sample several minutes after a drop of water has been applied to it. The THz phase of the transmission data is converted with the help of calibration data in Fig.3 to show the absolute water content as a function of position on the sample. The diffusion-induced spreading of the water in the paper is clearly identified. In the future, the temporal evolution of the moisture distribution could be monitored with real time THz-imaging systems.

For the paper manufacturer, it is interesting to estimate of the maximum paper weight measurable with the imaging setup. Heavier paper typically can be loaded with a proportionally larger amount of water. Based on the approximately linear dependence, we can simply scale up the strongest absorption of our measurements to the noise limit. The highest measured relative optical-density of about 1.5 (corresponds to 15 dB damping) is compared to the dynamic range of 60 dB (measured without sample, the low optical density of dry paper is negligible here) at a lock-in integration time of 10 ms [17]. Assuming the thickness of the paper and the saturation areal density of water are both proportional to the paper weight, the total (bulk) optical-density of a saturated paper sample will also be proportional to the paper weight. Hence, four times the absorption of a $80\,\mathrm{g/m^2}$ sample with $\approx$ 2.5kg water/kg fiber would attenuate the signal down to the noise level. So for high moisture content the upper limit estimation is about $320\,\mathrm{g/m^2}$.

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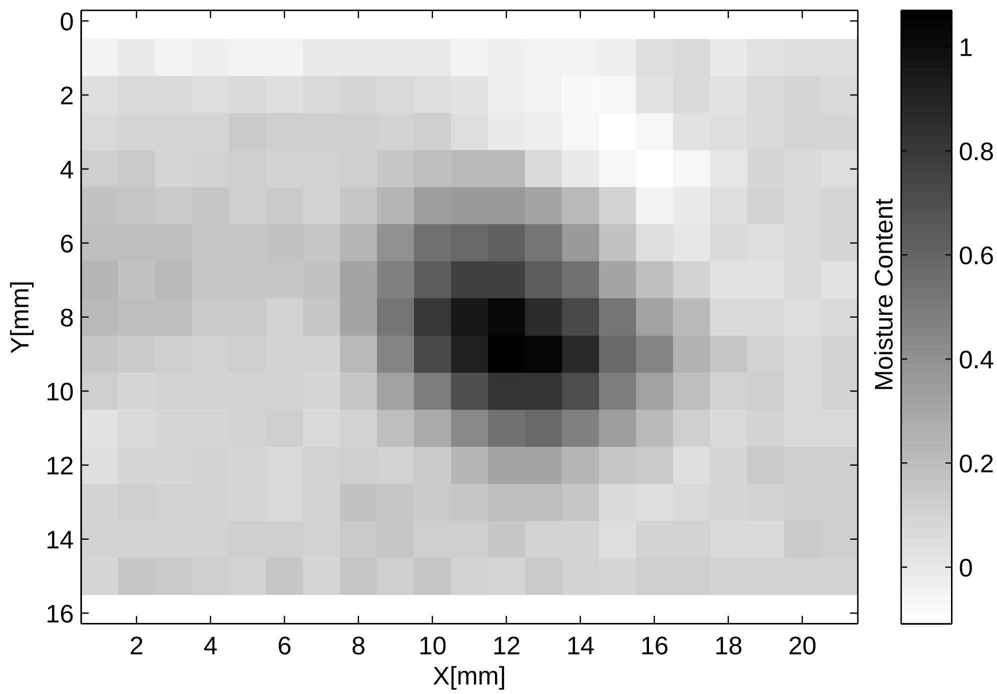

Fig. 4. Transmission image through $80\,\mathrm{g/m^2}$ paper at 0.6 THz showing the distribution of water in terms of moisture content (absolute values of kg water per kg fiber)

This suggests that measurements with THz radiation can not only cover a wider range in terms of moisture content compared to prevailing NIR meters, but also should cover a wider range in terms of paper weight (due to the lower residual absorption/scattering of paper in the THz range).

4. Conclusions

In summary, we have explored the potential of THz transmission spectroscopy to quantitatively determine the amount of water in paper. Working at a frequency of 0.6 THz, we find that both amplitude and phase measurements allow to accurately quantify the moisture content. At this frequency, scattering of the radiation at the surface of paper is weak and the spatial resolution can be in the sub-millimeter range. These findings make THz imaging systems attractive as a possible tool for online process monitoring in paper fabrication.

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