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THz spectroscopy techniques for water gradient quantification

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Abstract—Understanding depth dependent hydration, or gradients, is important for a wide range of applications from paper manufacturing to biological tissue imaging. However, in situ characterization, especially in a non-destructive, reflective measurement is difficult in practice. Moreover, uncontrolled sample drying, water migration, and surface warping create significant challenges towards experimental testing. In this work we show a simple scenario with a standard cleanroom wipe of around 350 μm thickness being exposed to water and left for complete drying, while its reflectivity together with changing mass data were recorded. The reflectivity was measured using VNA-based quasioptical system. High dynamic range, frequency domain measurements show a non-trivial frequency dependence on reflectivity that cannot be explained by aqueous dispersion alone. Further, the measurements reveal complicated drying dynamics where broadband reflectionless behavior was observed even when the sample was still ~ 50% water by mass. These investigations are an important step towards model model-free based analysis of spectra that should provide direct extraction of water at depth.

I. INTRODUCTION

Accurate quantification of water content and hydration gradients play a key role in a range of fields from medical imaging to paper production[1], [2], [3], [4]. Conventional biophotonics is not typically applied to the spatiotemporal mapping of tissue water content. The high sensitivity of THz waves to the water content and its state in tissue allows application of this frequency range to medical diagnosing, paying particular attention to the contrast of the image observed between healthy and pathological tissues.

The majority of in situ water gradient analysis is model-based (e.g. Fick’s Law) where the hydration at varying depths is determined by the sample boundary conditions. This approach has achieved significant success in varying tissue types. However, it’s possible that some phantoms, especially under low water content conditions or involving sample matrices with unknown water transport mechanisms, may deviate from these assumptions. In this case, a model free based analysis where the water content at any particular depth can be directly sensed, would aid in the understanding of spectra acquired from complex aqueous targets.

In this work, we report new experimental results of concomitant measurements of sample weight and THz reflectivity over ~ 90 minutes. Improvements in experimental protocol and data analysis compared to [5] revealed previously unseen dynamics where reflectionless behavior corresponded to significant remaining water content. Additionally, Optical Coherence Tomography and THz TDs measurements of the samples were performed in attempt to link drying behavior to surface roughness and fiber density.

II. METHODOLOGY

In our study, we assume that a piece of wet cleanroom wipe left for drying in natural conditions with constant temperature and humidity, is drying gradually from top to bottom. The drying process that involves gravity together with wipe properties and composition, creates a time-dependent gradient. To ensure planarity and absence of air gaps, we fixed the sample on a thick (25mm) polypropylene (PP) brick using the clips, like it is shown in Figure 1 (c). The sample holder is placed on the precision balance Radwag RA-PS 450.3Y to control the change of mass during the drying process, Figure 1 (d). Together with mass, the change in the sample’s THz reflectivity was recorded over time. The reflectivity measurements by means of S₂₁ parameter were taken with the vector network analyzer (N5225B PNA by Keysight Technologies) coupled with TxRx (transmitter/receiver) modules operating at 325-500 GHz range (WR-2.2 VNAX by Virginia Diodes Inc.)

Fig. 1. Experimental setup. Panel a illustrates the photograph of the actual setup, b is a schematic representation, c the photograph of the sample holder (PP brick) with the sample, and d is the schematic representation of the sample holder setup.

We want to clarify the possible misunderstanding of terms: S₂₁ is the transmission coefficient, however, using a designed setup we record the signal going from one TxRx module, next reflected by the sample, and next guided to the other TxRx module via a set of optics, so we call it “reflection” even though it is expressed with the S₂₁.

The refractive indexes of the PP sample holder and cleanroom wipe are close but not equal, therefore we have a layer having 85% water by volume which is bounded by water from one side, and air from the other side. This notion of the little difference between the refractive indexes of studied layers is important since we want to ensure the detection and sensing capabilities even when the contrast between the layers is low.
An optimum measurement procedure has been developed after a series of trials and it was set as follows: at the beginning the sample is dry and we record its mass together with THz reflectivity. Approximately one minute after the start of the experiment, 230μl of water was added to the wipe. This amount of water for the selected wipe size ensures that the sample is saturated with no extra water being leaked outside.

To visually observe the possible change in the sample thickness, fiber structuring, or presence of air gaps between the sample and the PP holder after water deposition, we took the OCT images every minute during the drying process. We observed, that immediately after water was added, the wipe swells and gets thicker, and we also saw a clear contrast difference between the wet wipe and PP, see Figure 2. That is related to the change in the composite’s refractive index. From the OCT images, we also see that the fibers on the outer surface of the wipe were pressed by the water and remained to be flatter after drying, compared to the initial dry state.

The top and bottom surfaces of the standard cleanroom wipe made of Polyester (45%)/Cellulose (55%) mix are not structurally equal: one is smoother than the other. In fact, the fibers of the wipe are more compacted on one of the sides. This structural difference leads to a different water suction, and therefore, to the drying process. For clarity we name these two cases as side A, being the rough, fluffy side, and side B which is the smooth side, respectively.

To learn more about samples, we did additional study using the THz TDS setup from TeraView, in a configuration shown on Figure 3. The measurement protocol kept the same as for the VNA-based measurements.

### III. Results

Obtained TDs data provide us with some more explanation. The result showed the influence of fibers constitution/packaging within the sample. Now we see that the waveforms of the samples for “Side A” and “Side B” are different accordingly. First, we measured dry samples at time zero: Figure 4 (a), and next time-dependent THz TDs waveforms for sides A and B individually: (b) and (c) figures, respectively.

![Fig. 2. OCT images of the sample (rough surface UP). Top left: dry sample at time 0 min. Top right: 225μl of water added, time 1 min. Bottom left: middle of the experiment, time 60 mins. Bottom right: water dried out, 115 min, end of the experiment.](image1)

![Dry samples](image2)

![Time, ps](image3)

![Side A](image4)

![Side B](image5)


The VNA based reflectivity measurements shows even more significant evidence of the difference for these two cases, see Figure 5.

Although recorded weight data shows the same drying tendency for sides A and B, the $S_{21}$ data differs a lot. For side A we observe two dips below the dry reflectivity state, while for side B there is only one, closer to the end of the experiment. Measurements were done tens of times showing this behavior repeatedly.

Fig. 4. THz TDs measurements. Panel a shows sides A and B at time zero, when the samples are dry; panel b shows the TD waveforms for the sample A at different time points during the drying process; panel c shows the TD waveforms for the sample B at different time moments during the drying process. Both samples were saturated with water at time 1 min, and at time 90 minutes they are completely dried out.

Fig. 5. Change of $S_{21}$ of the wet wipe left for drying in a natural environment. Side A indicates that the wipe is positioned with rough-structured fibers facing the air, while Side B is the opposite case: rough-structured fibers facing the PP holder.

The most interesting feature is the presence of the first absorption peak seen for side A. At the time point corresponding to the peak, the sample still has a significant amount of water. All the experiments described above have motivated us to put an effort on developing the theoretical tools to explain the observations.

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