Characterization of moisture in bitumen using continuous wave terahertz spectroscopy

Zhangyue Wei,[1] Lili Ma,[2] Alaa Jabbar Jumaah,[1] Idelfonso Tafur Monroy[1] and Min Wan[1*]

[1] Department of Electrical Engineering, Eindhoven University of Technology, Eindhoven, 5600 MB, Netherlands.
[2] Faculty of Civil Engineering and Geosciences, Delft University of Technology, Delft, Netherlands.

*Corresponding author: m.wan@tue.nl

Accurate measurement of moisture content in construction materials, such as bitumen, is crucial for various infrastructure applications. Current methods for detecting moisture content in bitumen have several drawbacks, such as long-term testing durations or the destruction of the sample's microstructure during measurement. Terahertz (THz) spectroscopy is a powerful tool in determining moisture content in materials rapidly and non-destructively. This study explores the application of THz spectroscopy in assessing moisture diffusion in bitumen, which impact its field performance and durability. Bitumen samples with varying moisture contents were prepared by soaking them in water for durations ranging from 0 to 8 days. All samples were subsequently analysed using a continuous-wave (CW) THz spectroscopic system in a transmission mode with the frequency range of 0.1 to 1.9 THz. Results from the THz measurements were analysed and compared with water diffusion model, providing comprehensive validation of the THz spectroscopy results. We demonstrate the capability of THz spectroscopy to provide precise moisture content in bitumen, which could be used to evaluate moisture damage of the bitumen in the future.

Introduction

Presence of moisture in construction materials leads to its deterioration, thus it is necessary to determine the moisture content to further evaluate the moisture damage. One of the construction materials is bitumen, a byproduct of oil refinery, which has been extensively used in asphalt pavements. Presence of moisture in bitumen significantly affects its durability [1]. Currently, the determination of moisture content in bitumen sample is both time-consuming and complex. One commonly employed method is the Dynamic Vapor Sorption (DVS) system, where the moisture content is determined by measuring the weight change in a material sample using a high resolution micro-balance. However, a typical DVS measurement can take several days to complete [2]. Alternative techniques, such as spectroscopy, have also been explored for moisture content evaluation. Fourier Transform Infrared (FTIR) spectroscopy, for instance, is used to assess moisture content. While FTIR offers rapid analysis, its limited penetration depth restricts its effectiveness to extremely thin samples, making it challenging to prepare samples with precisely controlled thickness [3].

The frequency range of Terahertz (THz) wave lies between infrared and microwaves, THz has higher penetration capabilities through non-conducting materials compared to infrared. This, together with the non-ionizing nature of the radiation, makes THz suitable for non-destructive testing. Since it is highly sensitive to water, thus has been widely used to examine water content level in biological samples [4].

In this paper, we investigate the moisture content in bitumen samples using continuous wave THz spectroscopy with a frequency range from 0.1 THz to 1.9 THz. The samples' moisture content is controlled by varying the days they are soaked in water. The water mass of the prepared sample has been estimated by Fick's second law of diffusion. The THz spectra of the bitumen samples has been analysed by principle component analysis (PCA). It shows that the simulated water mass has high correlation (R^2=0.995) with the calculated PCA value.

Method

A 70/100 penetration grade bitumen, subjected to pressure ageing vessel (PAV) conditions (2.1MPa and 100 ºC for 20 hours) to simulate 5-10 years of road service, was prepared, referred to as T710-1p. The sample was prepared by weighing a specific amount of bitumen and placing it in a glass petri dish. This petri dish was then heated up in an oven at 140 ºC for 5 min to form a film with a thickness of 1 mm. To evaluate the moisture content of the sample using THz spectroscopy, six T710-1p have been prepared, by soaking them in water for 0, 1, 2, 3, 6, 8 days, which has been labelled as Day0, Day1, Day2, Day3, Day6, Day8, respectively.

The measurement was performed with a wideband transmission CW THz system TeraScan 780 from TOPTICA®, with frequency from 0.1 to 1.9 THz and with a step size 40 MHz, as shown in Fig. 1a, Fig. 1b. Two tuneable 780 nm lasers are combined, and their output is coupled to a GaAs photomixer transmitter and receiver using optical fibers. The resulting THz wave is propagated through free space and transmitted through the sample under investigation. During the measurement, an empty glass petri dish was measured as reference. Each sample was carefully taken out from the water and the water around the petri dish and bitumen surface has been dried.

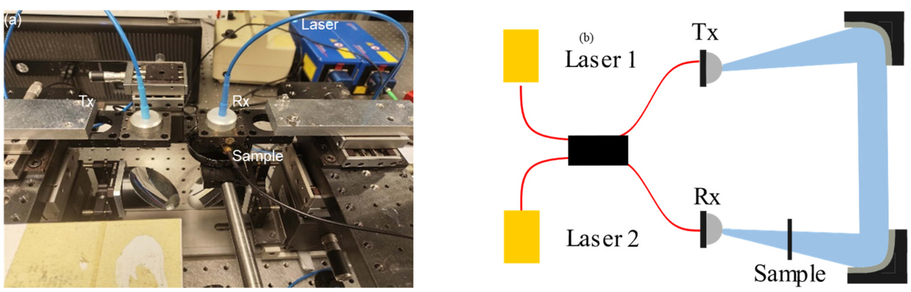

Figure 1. THz spectroscopy setup: (a) photo and (b) schematic of the system.

The moisture content in the sample is evaluated by estimating water mass, in the sample, predicted by Fick's second law of diffusion:

$$\frac{dC}{dt} = D\nabla^2 C$$

C is the concentration of water (mol/m^3), D is the diffusion coefficient (m^2/s), the numbers used in the equation are based on previous study which used the same material, the model achieves a root mean square error (RMSE) of 0.0025 [5]. After determining the concentration, the water mass is calculated by assuming the sample is a cylinder that has a radius of 2 cm and 1 mm height, which matches the dimension of T710-1p samples.

Result

The estimated change in water mass as calculated by Fick's second law of diffusion is shown in Fig. 2. Initially, there is a rapid increase in water mass within the samples, indicating swift moisture uptake. As time progresses, the rate of mass change converges as the water diffusion deaccelerates exponentially [5].

The obtained experimental spectrum data was processed using the Savitzky–Golay filter with a polynomial order of 4 and 25 points performed by MATLAB function *sgoloayfit t*o reduce signal oscillation [6]. The filtered THz spectra of six bitumen samples between 0.1- 1.9 THz are shown in Fig. 3a. A clear water absorption peak is visible around 0.56 THz, which can be clearly seen in the enlarged subfigure between 0.52 to 0.62 THz.

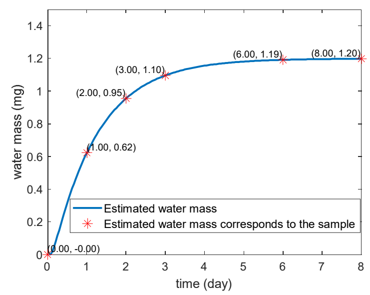

Fig 2. Estimated water mass in bitumen over time. The sample's corresponding time is highlighted with read star and their corresponding time and water mass.

Observing the enlarged figure and peak at 0.56 THz, it shows that the spectrum of the samples display increased absorption with the progression of different days during the experiment. However, day 0 and day 1 results are higher than the measured reference at 0.56 THz. The reason why reference value is lower than result from day 0 and day 1 is likely due to attenuation caused by the glass of the petri dish, which interferes the transmitted THz signal. Thus, high transparency material at THz frequency will be used for further study.

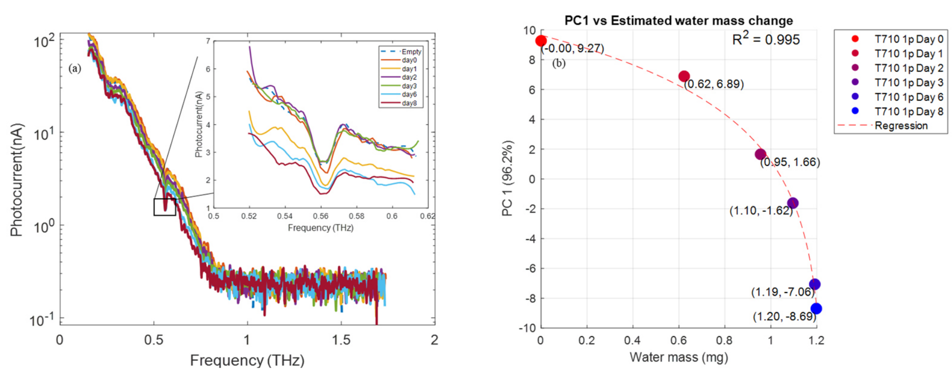

Fig 3. (a) The filtered THz spectra from 0-1.9 THz for T710-1p undergoing for 0, 1, 2, 3, 6, 8 days of water immersion. The enlarged figure shows the result from 0.52-0.62 THz. (b) PC1 vs Estimated water mass change and their corresponding water mass and PC1. The dash line is fitted with logarithmic regression.

THz spectral data between 0.52 to 0.62 THz were unified to the same size to perform PCA. The data has been transformed into a new set of uncorrelated variables called principal components [7]. The first principal component (PC1) represents the largest variance and primary variation in the spectrum, result shows that 96.2% of variance of the spectrum can be explained by PC1. Given that the only variation between samples was the duration of water immersion, therefore, PC1 indicates the water level change. Fig. 3b illustrates the correlation between the calculated PC1 of different samples and the estimated water mass shown in Fig. 2 in bitumen. A logarithmic regression model was applied to fit the data, yielding an R^2 value of 0.995. This indicates that PC1 result from the spectrum is highly correlated with the result estimated by the Fick's second law of diffusion.

Discussion

In this study, a THz CW system was used to examine the moisture content in bitumen samples. Due to relativity high penetration capabilities of THz spectroscopy, the procedure of measuring bitumen samples is much easier than that of FTIR [3]. Each measurement takes around 5 mins to perform the measurement, which is much faster than the procedure of DVS [2].

Result from the spectra of T710-1p samples revealed that the THz absorption intensity is affected by water concentration. The spectrum of the samples display increased absorption with the progression of days, attribute to a higher moisture content within these samples as predicted. In addition, the difference between the results on Day 6 and Day 8 is minimal, suggesting a slowdown in the diffusion process. This observation is consistent with result from previous study [5]. However, the reference measurement spectrum has lower value than that of the sample from day 0 and 1. Several factors might contribute to these discrepancies, e.g., the glass petri dish has thickness of 1.72 mm, which attenuate THz wave and the interaction with spherical waves passing through the glass could introduce muti-reflections in the measurement.

There are several limitations in this study: (a), the glass petri dish appears to be highly absorbing THz wave, thus it is ideal to use material that is more transparent to THz wave, such as Teflon or quartz; (b), future measurement should include more samples at the same day to check the consistency of the measurement; (c), the estimated water mass is based on assumption that the material has same diffusion as the previous study [7], thus in the future measurement, result from THz should be compared with measurement method that had been previously validated, such as DVS or FTIR.

Acknowledgement

This work is partly funded by the project 4TU RECENTRE (OA102070) and EWUU HOBO seed fund. The authors would like to acknowledge the support from Center for Terahertz Science and Technology Eindhoven.

Reference

[1] Kassem, E., Masad, E., Lytton, R., & Bulut, R. (2009). Measurements of the moisture diffusion coefficient of asphalt mixtures and its relationship to mixture composition. *International Journal of Pavement Engineering*, *10*(6), 389–399.

[2] Cheng, D., Little, D. N., Lytton, R. L., & Holste, J. C. (2003). Moisture Damage Evaluation of Asphalt Mixtures by Considering Both Moisture Diffusion and Repeated-Load Conditions. *Transportation Research Record: Journal of the Transportation Research Board*, *1832*(1), 42–49.

[3] Vasconcelos, K. L., Bhasin, A., & Little, D. N. (2010). Measurement of Water Diffusion in Asphalt Binders Using Fourier Transform Infrared–Attenuated Total Reflectance. *Transportation Research Record: Journal of the Transportation Research Board*, *2179*(1), 29–38.

[4] Nikitkina, A. I., Bikmulina, P. Y., Gafarova, E. R., Kosheleva, N. V., Efremov, Y. M., Bezrukov, E. A., Butnaru, D. V., Dolganova, I. N., Chernomyrdin, N. V., Cherkasova, O. P., Gavdush, A. A., & Timashev, P. S. (2021). Terahertz radiation and the skin: A review. *Journal of Biomedical Optics*, *26*(04).

[5] Ma, L., Varveri, A., Jing, R., Kasbergen, C., & Erkens, S. (2022). Thermodynamics and kinetics of moisture transport in bitumen. *Materials & Design*, *222*, 111028.

[6] Helal, S., Sarieddeen, H., Dahrouj, H., Al-Naffouri, T. Y., & Alouini, M. S. (2022). Signal Processing and Machine Learning Techniques for Terahertz Sensing: An Overview. *IEEE Signal Processing Magazine*, *39*(5), 42–62.

[7] Jolliffe, I. T., & Cadima, J. (2016). Principal component analysis: A review and recent developments. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, *374*(2065), 20150202.