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A Novel Method for the Deterioration State Evaluation of Mineral Insulating Oil by THz Time-Domain Spectroscopy

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ABSTRACT The operating life of power transformers is determined by the insulation state of the transformer insulating materials, which has been widely recognized. Herein, it is a new attempt to diagnose the state of the transformer mineral insulating oil by the terahertz time-domain spectroscopy (THz-TDS), taking advantage of its high signal-to-noise ratio and fast, accurate, and stable data acquisition characteristics. First, the THz time-domain waveforms of mineral insulating oils with different insulation states prepared by the accelerated thermal aging method were measured. Second, the Fabry–Perot (F-P) effect of instruments and container is eliminated using the improved electromagnetic wave transfer function. Then, the THz spectral parameters of insulating oils, such as absorption coefficient and refractive index, can be calculated. Meanwhile, the dielectric properties of insulating oils in the terahertz region are obtained. Finally, compared with traditional physicochemical diagnostic indicators, the results preliminarily prove that THz-TDS technology can effectively evaluate the state of the mineral insulating oil.

INDEX TERMS Mineral insulating oil, aging condition, THz time-domain spectroscopy (THz-TDS), spectral parameters, dielectric properties.

I. INTRODUCTION

The safe operation of power grid is the basis of ensuring stable and reliable power supply, in which power transformer plays a vital role [1]. Whether the transformer can operate safely depends largely on the integrity of the insulating components [2]. Therefore, the research on transformer internal insulation system has been highly valued by the power sector and experts. Mineral insulating oil is the main component of insulation system in power transformer. During the long-term operation of the transformer, the mineral insulating oil deteriorates and must be filtered or replaced. Due to the difference of actual operation and load of transformers, their residual operating life varies greatly. To avoid the economic loss caused by blind maintenance and replacement, it is necessary to use modern technology and analytical means to diagnose and evaluate the residual life of in-service transformers. Current diagnostic techniques include return voltage measurements [3], polarization and depolarization current [4], [5] and frequency domain spectroscopy [6]–[8].

In recently years, time-domain spectroscopy (TDS) is a young method for studying insulation materials. Herein, terahertz technology was used to diagnose the deterioration of mineral insulating oil. Terahertz (THz) radiation has wavelengths of 30 μm$\sim$3 mm, typically referred to as the frequencies from 100 GHz to 10 THz, which lies in the frequency gap between the infrared and microwaves [9], [10]. Many organic molecules have strong absorption in the terahertz region due to the rotation and vibration transition of the dipole. Therefore, it is possible to study the properties of materials by different absorption and reflection of terahertz pulses [11]. At present, the THz time-domain spectroscopy (THz-TDS) has been used to obtain the transmission and reflection spectra of substances in the terahertz region, which has been widely used in security [12], non-destructive testing [13], medical [14] and other fields. In particular, terahertz fingerprint peaks have become one of the most promising technologies for substance detection in the future [15], [16].

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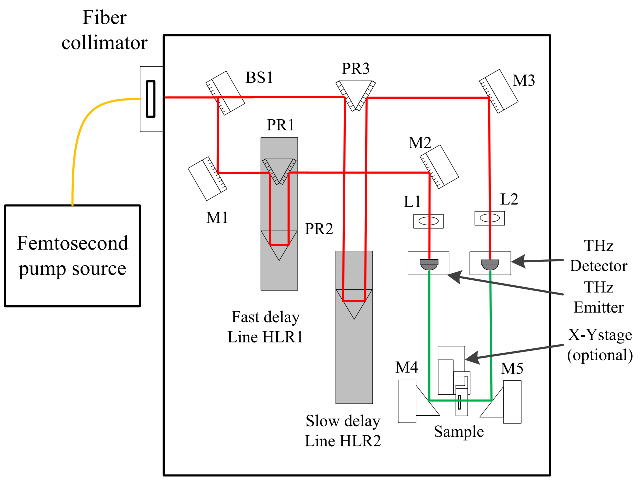

FIGURE 1. The optical layout of terahertz-time domain spectroscopy (THz-TDS) setup [19].

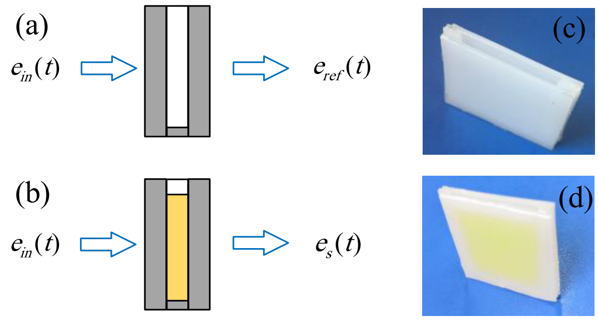

FIGURE 2. (a) and (b) Schematics and (c) and (d) photographs of the self-made PE container showing (a) and (c) empty container and (b) and (d) container filled with the sample.

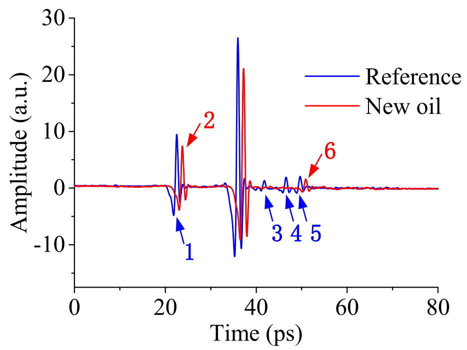

FIGURE 3. Time-domain spectrum of the reference and of new oil.

Currently, there are few reports on the use of terahertz technology to diagnose the degradation of insulating oil. Li *et al.* [17] used THz-TDS to detect the THz parameters of the insulating oil samples extracted from the in-service transformers in the range of 1.5-6.0 THz. However, the variation of optical parameters of transformer insulating oil during operation was not mentioned in the article. Kang *et al.* [18] used THz-TDS to detect the optical parameters of the pale yellow, bright yellow, amber, dark brown four-color transformer insulating oils in the range of 0.2-3.0 THz. But the disadvantage was that the color distinction was not objective enough. Herein, the variation of terahertz optical parameters of insulating oil in different operating periods is given, which solves the problems existing in current research.

II. EXPERIMENTS AND METHODS

A. SAMPLE PREPARATION

First, an accelerated thermal aging process was used to simulate the aged condition of transformer oil-paper systems to obtain insulating oil exhibiting different deterioration states. The oil used herein was 25# ordinary mineral oil produced in China (Karamay) and the paper was 0.5-mm-thick kraft paper produced by the Oriental Insulation Material Factory (Mianyang, China). A detailed description of the experimental steps can be found elsewhere [19]. Next, three types of insulating oil samples aged at 130°C for 3, 20 and 40 days were selected. Finally, the optical parameters in the low frequency terahertz range of 0.1–1.8 THz were tested, and the traditional discriminant method was introduced to assist in diagnosing the variation of terahertz optical parameters after insulating oil deterioration.

B. TERAHERTZ SPECTRAL ACQUISITION

The acquisition of the THz spectrum was carried out in a T-Spec THz spectroscopy system (Ekspla, Inc., Vilnius, Lithuania). The optical layout of THz-TDS system was shown in Figure 1. THz emitter and THz detector of the system consists of a micro strip antenna integrated with photoconductor and silicon lens mounted on the back side of photoconductive antenna. Low temperature grown GaBiAs was used as photoconductor. Total GaAs substrate thickness was about 500 μm. The pump laser can provide 1050±40 nm wavelength, 50-150 fs duration and more than 40mW output power. And the pulse repetition frequency was about 80MHz.

Owing to the weak absorption of a THz pulse by polyethylene (PE), the insulating oil samples were placed in a self-made 1-mm-thick polyethylene container (analytic grade >95%), shown schematically and by photographs in Figure 2. The signal of the empty container, $e_{ref}(t)$, was used as a reference. And in order to obtain a better signal-to-noise ratio (SNR), each spectrum was taken to the average of 1024 time-domain scans. The experiment was carried out at room temperature of 296 K, and relative humidity below 5% to avoid the effect of moisture.

C. DE-OSCILLATED PROCESSING

Figure 3. shows that, in addition to the primary pulse waveform, secondary peaks produced by multiple reflections were present in the spectra owing to the Fabry-Ferot (F-P) effect of the emitter and detector and the imperfect purity of the PE container. It can be clearly seen in Fig. 3 that four secondary peaks existed in the reference signal and two secondary peaks in the sample signal. The secondary peaks

of the reference signal are more than the sample, which may be due to the attenuation of multiple reflections in the sample. Unfortunately, these secondary peaks will cause a spurious spectral oscillation in the frequency domain, which may hide the important characteristics of the sample [20]. Therefore, an appropriate means must be chosen to eliminate these secondary peaks. Herein, the improved electromagnetic wave transfer equation was used to eliminate spurious oscillations of the frequency-domain spectrum, which does not reduce the frequency resolution nor require a specially manufactured device [21].

Herein, the calculation model proposed in references [22], [23] was used to eliminate the spurious oscillations caused by F-P effect of the containers and the emitter and detector. Frequency-domain function $E(\omega)$ can be obtained by fast Fourier transform (FFT) of measured time-domain waveform. $E_{ref}(\omega)$ and $E_s(\omega)$ were frequency-domain function of reference and sample waveforms, respectively. Then the transmission function $H(\omega)$ can be obtained, and was given as

$$H(\omega) = \frac{E_s(\omega)}{E_{ref}(\omega)} = \frac{4n_s(\omega)n_0}{[n_s(\omega)+n_0]^2} \cdot \exp\left[-\frac{\kappa_s(\omega)\omega d}{c}\right] \cdot \exp\left\{-j[n_s(\omega)-n_0]\frac{\omega d}{c}\right\} \quad (1)$$

where $n(\omega)$ and $\kappa_s(\omega)$ were complex refractive index and extinction coefficient, ω, c, d and n_0 were frequency, light speed, the thickness of the sample and refractive index of nitrogen, respectively.

For the reference signal $e_{ref}(t)$, the electromagnetic wave transfer equation was modified owing to the F-P effect, and was given as

$$\begin{aligned} e_{ref}(t) &= e_0(t) * \sum_{i=0}^{\infty} [a_i\delta(t_i) * h_i(t)] \\ &= e_{r0}(t) * \{a_0\delta(t_0) + \sum_{i=1}^{\infty} [a_i\delta(t_i) * h_i(t)]\} \end{aligned} \quad (2)$$

$$E_{ref}(\omega) = E_{r0}(\omega)\{1 + \sum_{i=1}^{n} [(a_i e^{-j\omega(t_i-t_0)})H_i(\omega)]\} \quad (3)$$

where $e_{r0}(t)$ is the primary THz pulse waveform; a_0 and t_0 are the amplitude and peak time of primary pulse signal, respectively; a_i and t_i are the amplitude and peak time of the secondary peak, respectively; and $h_i(t)$ is the transfer function of the absorption of the i-th oscillation. During the analysis, the time-domain signal was normalized so that $a_0 = 1$.

Similarly, the signal of the sample can be modified to

$$\begin{aligned} e_s(t) &= e_0(t) * \sum_{i=0}^{\infty} [a_i\delta(t_i) * h_i(t) * h_s(t)] \\ &= e_{s0}(t) * \{\delta(t_0') + \eta \sum_{i=1}^{\infty} [a_i\delta(t_i) * h_i(t) * h_s(t)]\} \end{aligned} \quad (4)$$

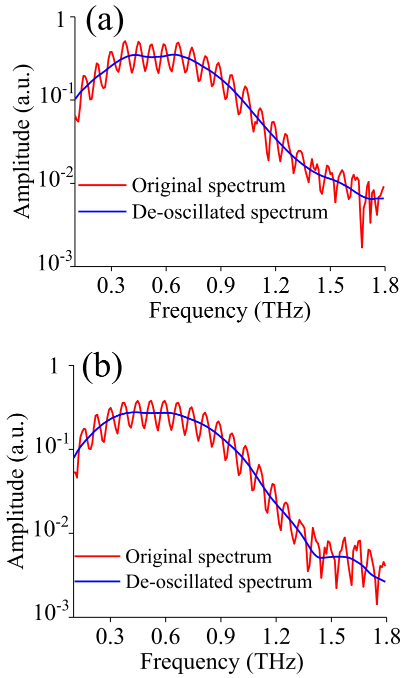

FIGURE 4. Comparison of original and de-oscillated spectra of the (a) reference and (b) sample signals.

$$\begin{aligned} E_s(\omega) &= E_{s0}(\omega)\{1 + \eta \sum_{i=1}^{2} [(a_i e^{-j\omega(t_i-t')})H_i(\omega)H_s(\omega)]\} \\ &= E_{s0}(\omega)\{1+\eta e^{j\omega(t'-t_0)} \sum_{i=1}^{2} [(a_i e^{-j\omega(t_i-t_0)})H_i(\omega)H_s(\omega)]\} \end{aligned} \quad (5)$$

where $e_{s0}(t)$ and t_0' are the primary pulse waveform and the peak time of the primary pulse of the sample, respectively; $h_s(t)$ is the projection function of the THz pulse passing through the sample; and η is the modified coefficient equal to the ratio of the peak amplitude of $e_{ref}(t)$ to $e_s(t)$. The original and de-oscillated frequency-domain spectra of the reference and sample are shown in Figs. 4(a) and 4(b), respectively.

D. DATA PROCESSING

The Dorney *et al.* [24] and Duvillaret *et al.* [25] mathematical model was used to calculate the THz optical parameters of insulating oil, and was given as

$$n_s(\omega) = n_0 - \frac{\omega d}{c}\angle H(\omega) \quad (6)$$

$$\kappa_s(\omega) = \frac{c}{\omega d}\left\{\ln[\frac{4n_s(\omega)n_0}{|H(\omega)|\,[n_s(\omega)+n_0]^2}]\right\} \quad (7)$$

$$\alpha_s(\omega) = \frac{2\omega\kappa_s(\omega)}{c} = \frac{2}{d}\left\{\ln[\frac{4n_s(\omega)n_0}{|H(\omega)|\,[n_s(\omega)+n_0]^2}]\right\} \quad (8)$$

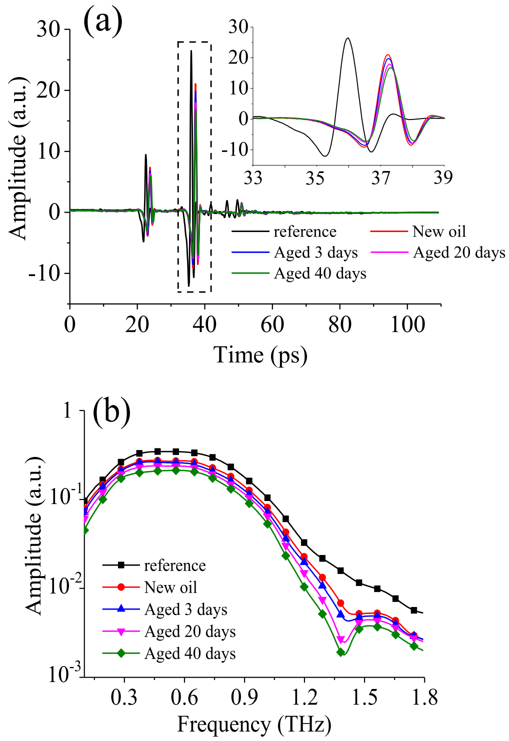

FIGURE 5. THz spectrum of reference and insulating oils with different aging time, (a) time-domain waveforms, (b) frequency-domain spectroscopy.

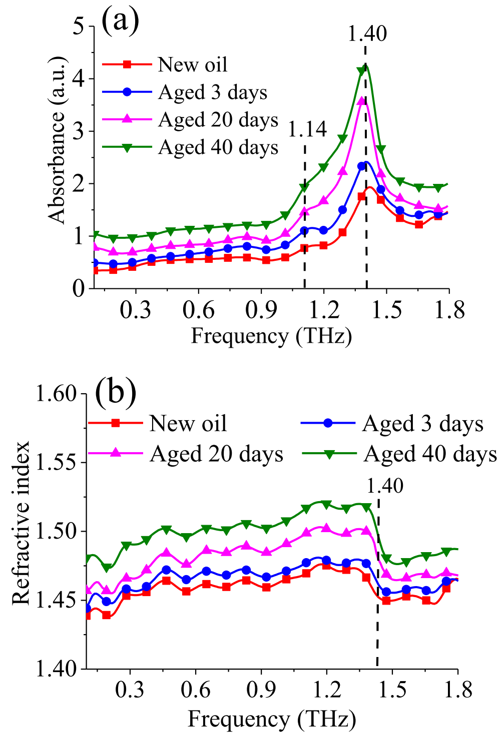

FIGURE 6. Spectral parameters of insulating oils with varying aging condition showing (a) absorption coefficient and (b) refractive index.

III. RESULTS AND DISCUSSIONS

A. TERAHERTZ TIME-DOMAIN WAVEFORMS AND FREQUENCY-DOMAIN SPECTRA

The THz time-domain waveforms (Fig. 5(a)) and de-oscillated frequency-domain spectra (Fig. 5(b)) of the reference signal and insulating oil with different aging conditions are shown in Fig. 5. It can be seen that the spectral amplitude of the insulating oil was attenuated and the time was delayed compared to the reference spectra, where longer aging times corresponded to longer time delays and more severe peak attenuation. These results are because the THz pulse was absorbed, reflected, and scattered on the surface of the insulating oil, causing peak attenuation. Further, the THz pulse has different propagation speeds in the air and oil samples, and thus the oil samples with various aging times exhibited different time delays compared to the reference. In the de-oscillated frequency-domain spectra, spectral pits appeared in the frequency amplitude of the oil spectra that were not present in the reference spectra. Further, longer aging times induced larger spectral pits, and thus the presence of these pits can be assigned as the THz optical characteristic of the insulating oils.

B. ANALYSIS OF ABSORPTION AND REFRACTION CHARACTERISTICS

Figure 6 shows the absorption coefficient and refractive indices of the transformer insulating oil with varying thermal aging condition in the range of 0.1–1.8 THz, which were calculated based on the frequency-dependent formulas. Figure 6(a) reveals that the baseline of the absorption coefficient spectrum increases with the frequency, which may be owing to the higher frequency enabling stronger absorption and scattering of the THz wave. It can be easy found that the insulating oils have a weak absorption peak at 1.14 THz and a strong absorption peak at 1.40 THz. Furthermore, the absorption becomes stronger with increasing aging condition, which may be owing to the increased amount and severity of the physical and chemical reactions. And the more severe the aging of insulating pressboards, the higher the mobility of polar substances such as water and small molecule acids in pressboards to oil, resulting in the greater polarity of insulating oil. Therefore, the more serious the aging condition is, the stronger the absorption of THz wave by insulating oil is. So, these THz absorption peaks can be assigned as characteristic fingerprints of mineral insulating oil degradation. Figure 6(b) plots the average refractive indices of the insulating oils in the range of 0.1-1.8 THz, which for the new mineral oil and that aged 3, 20, 40 days were found to be 1.459, 1.466, 1.478, and 1.498, respectively. These values reflect the absorption and dispersion characteristics of the insulating oils in various states, and exhibit an obvious change in the refractive index at the location of the strong characteristic absorption peak. This phenomenon indicates that the change properties of the refractive index can assist in determining the true absorption peak position of some substances [26].

As the operating life of the transformer increases, the insulating pressboards will deteriorate to produce 2-furaldehyde,

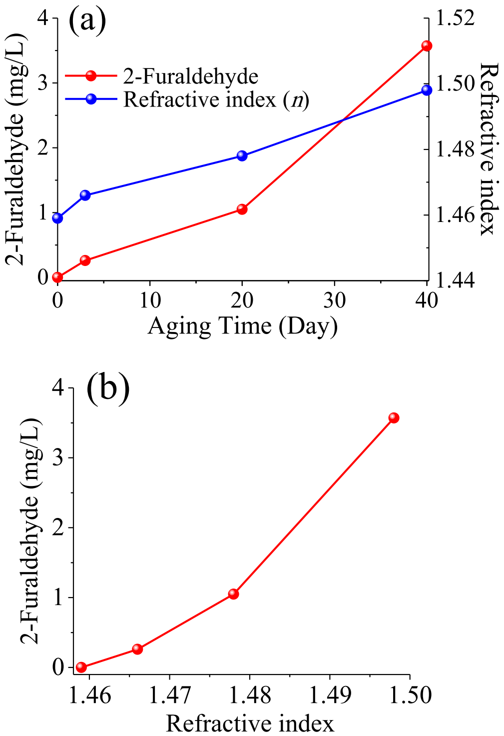

FIGURE 7. (a) Content of 2-furaldehyde in and refractive index of the mineral insulating oil vs. aging time and (b) 2-furaldehyde content vs. refractive index.

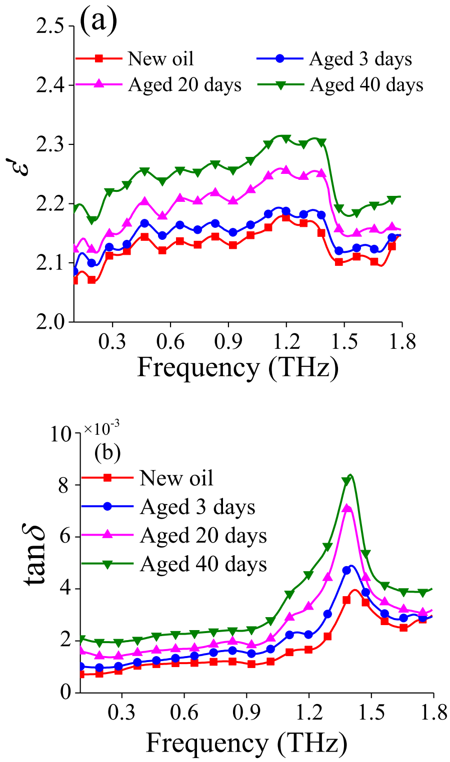

FIGURE 8. Dielectric constant of insulating oil in the THz region for oil aged for various times; (a) real part and (b) imaginary part.

which will be dissolved in the insulating oil. Therefore, the content of 2-furaldehyde is widely used as an important indicator for evaluating the residual life of insulating oil [27]. Figure 7(a) plots the 2-furaldehyde content in insulating oil and the refractive index of the oil as a function of aging time, where the 2-furaldehyde content was measured by high-performance liquid chromatography. It is seen in Fig. 7(a) that the refractive index has the same trend as the 2-furaldehyde content. Figure 7(b) plots the 2-furaldehyde content vs. refractive index, which indicates that the THz refractive index can be used as an effective indicator for the diagnosis of insulating oil deterioration.

C. DIELECTRIC PROPERTIES IN THE TERAHERTZ REGION

Currently, the state estimation method of insulating oil-paper based on the dielectric response theory has attracted wide attention [28], [29]. Dielectric response refers to the polarization and relaxation of dielectrics under an external electric field. As a typical composite dielectric, mineral insulating oil undergoes oxidation and hydrogenation during operation, breaking down the C–H and C–C bonds to ultimately produce gases such as CH_4 and C_2H_2, which will affect its dielectric properties. Therefore, it is necessary to know the dielectric properties of the transformer insulating oil.

Because THz-TDS can simultaneously obtain the amplitude and phase information of the sample, its dielectric properties can be easily obtained. According to the mathematical in references [30], [31], the dielectric constant in the THz region can be obtained by

$$\varepsilon(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega) = (n(\omega) + i\kappa(\omega))^2 \quad (9)$$

$$\varepsilon'(\omega) = n^2(\omega) - \kappa(\omega)^2 = n^2(\omega) - \frac{\alpha^2(\omega)c^2}{4\omega^2} \quad (10)$$

$$\varepsilon''(\omega) = 2n(\omega)\kappa(\omega) = \frac{\alpha(\omega)n(\omega)c}{\omega} \quad (11)$$

$$\tan\delta = \frac{\varepsilon''(\omega)}{\varepsilon'(\omega)} = \frac{4\omega c\alpha(\omega)n(\omega)}{4\omega^2 n^2(\omega) - c^2\alpha^2(\omega)} \quad (12)$$

where the resulting dielectric constant values (Fig. 8(a)) and the dielectric loss tangent ($\tan\delta$) of the insulating oil (Fig. 8(b)) are plotted in Fig. 8. The extinction coefficient was much smaller than the refractive index value, and so the real part of the sample dielectric constant was almost equal to the square of the refractive index. Since the refractive index of the sample changes by a small amount, and the frequency and speed of light are constant, the trend of the dielectric loss tangent ($\tan\delta$) is similar to the absorption coefficient. With increasing thermal aging time, the dielectric constant and the dielectric loss tangent ($\tan\delta$) increased, which is consistent with existing research [32], and thus the dielectric constant in THz region can indirectly indicate the condition of the insulating oil.

IV. CONCLUSION

In summary, this paper reports a novel method for evaluating the deterioration state of mineral insulating oil using THz-TDS technology. insulating oils with different conditions were obtained by an accelerated thermal aging method. Then the THz waveforms of the insulating oils were measured, and the spurious oscillations of frequency-domain spectra were eliminated using the improved electromagnetic wave transfer function. Finally, the THz optical parameters, such as the absorption coefficient and refractive index, of the insulating oils with varying condition were obtained. The mineral insulating oil was found to exhibit two absorption peaks at 1.14 and 1.40 THz in the range of 0.1–1.8 THz. With increasing deterioration, the absorption peak intensity and refractive index were found to increase. Finally, the 2-furaldehyde content in the insulating oil was measured by high-performance liquid chromatography, which is widely used to evaluate the residual life of insulating oil. The results preliminarily showed that THz-TDS technology can be used as a novel method to evaluate the deterioration of transformer insulating oil. Specifically, the dielectric constants of insulating oil in the THz region were found to indirectly indicate the condition of the insulating oil.

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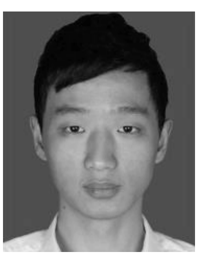

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