

Received April 27, 2019, accepted May 16, 2019, date of publication May 30, 2019, date of current version June 18, 2019.

Digital Object Identifier 10.1109/ACCESS.2019.2919969

# Significantly Improved Electrical Breakdown Strength of Natural Ester Liquid Dielectrics by Doping Ultraviolet Absorbing Molecules

**SUNING LIANG<sup>1</sup>, FEIPENG WANG<sup>1</sup>, (Member, IEEE), ZHENGYONG HUANG<sup>1,2</sup>, WEIGEN CHEN<sup>1</sup>, YOYUAN WANG<sup>1</sup>, AND JIAN LI<sup>1</sup>, (Senior Member, IEEE)**

<sup>1</sup>State Key Laboratory of Power Transmission Equipment & System Security and New Technology, School of Electrical Engineering, Chongqing University, Chongqing 400044, China

<sup>2</sup>Postdoctoral Research Station on Chemical Engineering and Technology, Chongqing University, Chongqing 400044, China

Corresponding authors: Feipeng Wang (fpwang@cqu.edu.cn) and Jian Li (lijian@cqu.edu.cn)

This work was supported in part by the National Natural Science Foundation of China under Grant 51425702, and in part by the Project 111 of the Ministry of Education China under Grant B08036.

**ABSTRACT** In this paper, we experimentally demonstrate that molecules with maximum absorption wavelength corresponding to the first excitation energy of base liquid molecules are able to significantly improve the electrical performances of natural ester oils. The positive lightning impulse breakdown and acceleration characteristics, ac breakdown characteristics, and streamer structure of the natural ester oils, as well as the electronic properties of the molecules, are presented. The molecules with different absorptions in ultraviolet lights were doped into the natural ester oil. The positive lightning impulse breakdown characteristics of natural ester oil with different doped molecules indicated that the doped molecule with great absorption in the medium wavelength ultraviolet irradiation was able to significantly enhance the positive lightning impulse breakdown and acceleration voltages of natural ester oil by 15% and 60%, respectively. In addition, it also enhanced the ac breakdown voltage by 17%. The base liquid molecules exhibited first excitation energy of 4 eV, which corresponds to the maximum absorption wavelength of such molecules, indicating that the improved electric breakdown strength was a result of the reduced the number of excited molecules and weakened ionization of molecules during the streamer propagation.

**INDEX TERMS** Natural ester liquids, lightning impulse dielectric properties, ac breakdown characteristics, molecular properties, streamer, ultraviolet absorbing molecule, first excitation energy.

## I. INTRODUCTION

The fire point of the natural ester (NE) liquid dielectrics (ca. 360°C) was more than 200°C higher than that of mineral insulation (MI) oil (ca. 160°C). This distinctive property indicates the potential application of NE oil in power equipment in heavily populated areas such as urban communities, which require high-level safety regulations. NE oil is 95% biodegradable within 28 days under natural conditions, indicating its applicability as a substitute for MI oil [1]. NE oil has more than 20% overload capacity, has a considerably lower life-cycle cost, and is rendering the expanding applications in distribution transformers [2]. Currently, NE oils are applied

The associate editor coordinating the review of this manuscript and approving it for publication was Debasish De.

to high-voltage power transformers in several startup cases at varying voltages [3].

The lightning impulse (LI) breakdown voltages  $V_{b-LI}$  of NE and MI oils are comparable in short oil gaps no longer than 25 mm [4]. However, for longer oil gaps, the  $V_{b-LI}$  for NE oil tends to show increasingly lower values than those of MI oil. In order to recognize the dynamic process before the arc breakdown which is typically exhibited with streamers, the acceleration voltage  $V_{a-LI}$  is introduced to determine the critical voltage upon which a streamer develops by average propagation velocity of above 10 km/s [5]. NE oil is prone to generate fast streamers at a much lower acceleration voltage  $V_{a-LI}$  than that of MI oil. This behavior has become more typical in severe non-uniform electric fields with an oil gap longer than 50 mm [6]. Fast streamers are confirmed as one of the most risky factors in producing insulation failures in

power transformers [7]. Due to the polarity effect, positive streamers tend to propagate faster than negative ones, which constitutes a greater risk of insulation failure [8]. This has promoted us to employ positive LI voltages for electrical breakdown measurements.

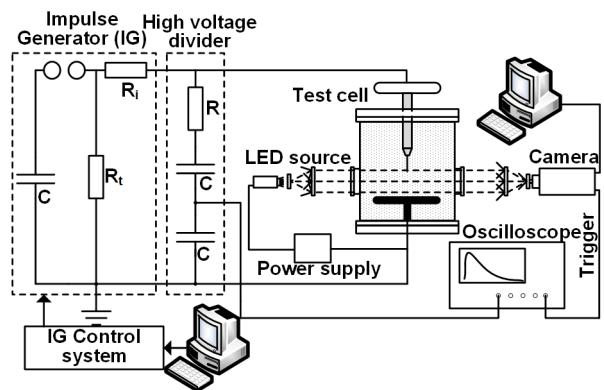
Some additives such as nano-particles and low ionization potentials (IP) additives were used to improve the breakdown characteristics of NE oil [9]. Li et al. observed enhanced LI and AC breakdown voltage following the addition of conducting nanoparticles  $\text{Fe}_3\text{O}_4$  [10]. Zhong et al. found that semiconducting nanoparticles,  $\text{TiO}_2$ , enhanced the AC breakdown voltage ( $V_{b-\text{AC}}$ ) of ester based nanofluids [11]. However, the long-term dispersion stability of nanofluids is quite poor under electrical, thermal, and magnetic stresses. Unge et al. observed that high-oleic natural ester oil added with two types of low IP additives (N,N-dimethylaniline and azobenzenes) exhibited improved positive  $V_{b-\text{LI}}$  and  $V_{a-\text{LI}}$  [12]. However, the both additives are highly toxic chemicals, which makes them impossible for real applications. NE oils, composed of triglyceride molecules, contain a lot of carbonyl groups and carbon-carbon double bonds vulnerable to molecule breaks under the ultraviolet radiation. The enhancement in electrical breakdown strength of NE oil is rarely reported by ionization restriction with ultraviolet absorbing molecules, whose maximum absorption wavelength correspond to the first excitation energy of base liquid molecules.

In this paper, we aim to demonstrate a route for improving the LI and AC breakdown behavior in NE oil by doping molecules with absorption in ultraviolet lights because of the expected ionization restriction. Molecules with different absorption in ultraviolet lights were doped into the NE oil. The LI breakdown characteristics of NE oil with different doped molecules indicated that the doped molecule with great absorption in the medium wavelength ultraviolet irradiation was able to significantly enhance the electrical performances in LI breakdown, acceleration and AC breakdown voltages. In order to have a deeper understanding of the effect of the doped molecule on the streamer in NE oil, the shadowgraphic images of streamers were recorded and analyzed. The doped molecule could inhibit the propagation of the streamer in the NE oils. For deeper molecular level insights into the mechanism, the relevant molecular properties of the molecules used in the experiments were calculated by Gaussian software. The molecules with maximum absorption wavelength corresponding to the first excitation energy of base liquid molecules could significantly improve the electrical performances of NE oils.

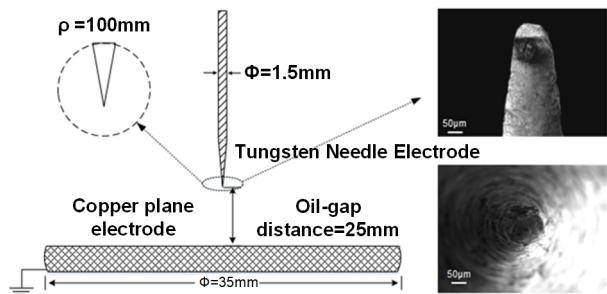
## II. EXPERIMENTAL SETUP

### A. THE SYNCHRONIZED MEASUREMENT SYSTEM AND ELECTRODES

The schematic diagram of the test system was shown in Fig. 1. The test system was made up of three parts-LI voltage generator and measurement system of applied LI voltage, self-designed test cell and shadowgraphic imaging system. A cuboid test cell with two quartz glass observation windows



**FIGURE 1.** The setup for streamer imaging and breakdown testing under LI voltages.

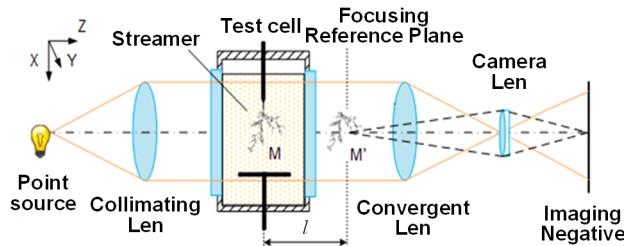


**FIGURE 2.** The structure and parameters of needle-plane electrode.

was used to hold the oil samples. A 6-stage Marx impulse generator with a maximum voltage of 900 kV was used to provide the standard LI voltage  $1.2 (\pm 30\%)/50 \mu\text{s} (\pm 20\%)$ . The waveform of applied voltage was recorded by the oscilloscope (LeCory WaveRunner 610zi) with a capacitance resistance divider of 891 times attenuation. The oscilloscope could synchronously output a trigger signal to high-speed camera (Photron SA5). The streamer propagation was examined by a shadowgraphic imaging system, which can help us to research effects of additives on pre-breakdown phenomena in the liquids under LI voltage [13]. The high speed camera frame rate was 10,000 frames per second and the exposure time of image was  $100 \mu\text{s}$ . The needle-plane electrode (Fig. 2) was used to simulate the severe non-uniform electric field under LI voltage. The needle-plane electrode configuration was consisted by a tungsten needle and a copper plane electrode with diameter of 35 mm. The distance of oil gap between needle and plane electrodes was set as 25 mm. The curvature radius of the tungsten needle that could guarantee the stability of the tip structure after several discharges was  $100 \pm 10 \mu\text{m}$ .

### B. THE SHADOWGRAPHIC IMAGING SYSTEM

In view of the rapid development of streamers and the short exposure time of photographic records, more light energy is needed on the unit area to increase the photosensitivity of pictures. The focus shadowgraphic imaging system is often used to reduce the photographic record frame and increase the exposure of each point on the photo to obtain a clear



**FIGURE 3.** Principle diagram of shadow graphic system for streamer imaging.

image. The principle diagram of the focus shadowgraphic imaging light path is shown in Fig. 3. The light routing is composed of point light source, collimating lens, converging lens, camera lens and corresponding negative film. The camera lens focus on the reference focusing plane  $M'$  and the distance between actual discharge position  $M$  is  $l$ . The streamer under LI voltage change the refractive index of the liquid. With the development of the streamer, the refraction zone formed in the channel can deflect the background beam passing through it, thereby forming a streamer-like shadow with a bright background at the reference focusing plane  $M'$ . The streamer-like shadows were recorded by a high-speed CMOS device. The relative brightness variation of the final imaging has the following relationship with the optical path parameters:

$$\frac{\Delta I}{I} = l \int_{\zeta_1}^{\zeta_2} \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) (\ln n) dz \quad (1)$$

where  $I$  represents the light intensity,  $l$  is the distance between the focusing reference plane and the actual streamer,  $z$ -axis is the direction of light propagation, and the  $xy$  plane is parallel to the corresponding negative film.  $n(x, y, z)$  represents the refractive index of streamer,  $\Delta I/I(x, y)$  represents the obtained signal,  $\zeta_1$  and  $\zeta_2$  represent the corresponding propagation coordinates of the entrance and exit light, respectively.

### C. OIL SAMPLES AND EXPERIMENT METHOD

The NE oil used in the test was a commercial NE oil FR3 (made from soybeans by Cargill, USA) transformer oil. It was dried and degassed in a vacuum oil purifier drying oven at 75°C for 6 h, and its moisture content was below 30 ppm after processing. Two molecules with different absorption in ultraviolet lights were used in the test. One is the molecule with absorption in the medium wavelength ultraviolet irradiation 4-ethoxycarbonylphenyl-n-methyl-n-phenylformamidine (UV1). The other is the molecule with absorption in long wavelength ultraviolet irradiation 2-(4'-diethylamino-2'-hydroxybenzoyl) benzoic acid hexyl ester (UVA Plus). The molecules were doped into the NE oil by ultrasonic with medium agitation at 75°C for 30 minutes. Then the oil samples were dried and degassed in a vacuum drying oven at 75°C for 48 h. The oil samples were prepared with additive mass fraction increasing from 0% to 5%. The rising-voltage method [14] and the overvoltage test method were utilized to determine the  $V_{b-LI}$  and  $V_{a-LI}$ , respectively.

The average velocity of the streamer was obtained by dividing the oil gap distance over the breakdown time  $t_b$  when the applied voltage was higher than  $V_{b-LI}$ . The AC breakdown voltages were tested by IEC60156.

### D. ABSORPTION SPECTRA AND ELECTRONIC PROPERTY OF MOLECULES

The ultraviolet-visible (UV-Vis) absorption spectra of UV1 and UVA Plus were obtained using a Shimadzu UV 3600 spectrophotometer with H<sub>2</sub>O as a reference. To avoid the influence of the NE oil on the ultraviolet absorption spectrum, ethanol was used for the solution. Due to the UV1 and UVA Plus have a strong absorption capacity of ultraviolet light, the content of additives was 10 ppm to avoid exceeding the range of measuring instruments.

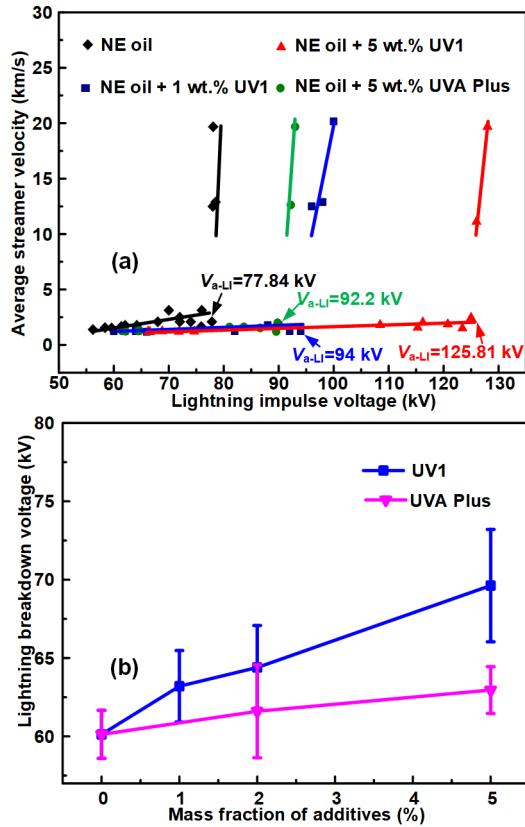
The electronic properties of molecules calculations were performed in the Gaussian 09 program package. The geometry of the molecules was optimized using density functional theory with Becke's three-parameter hybrid functional combined with the electron-correlation functional of Lee, Yang, and Parr (B3LYP) prescription employing the 6-311+G(d) basis set [15]. The IP and electron affinity (EA) were calculated as described in Ref. 16. The first singlet-singlet excitation energies are calculated by time-dependent density functional theory method with the same functional and basis set [17].

## III. EXPERIMENTAL RESULTS

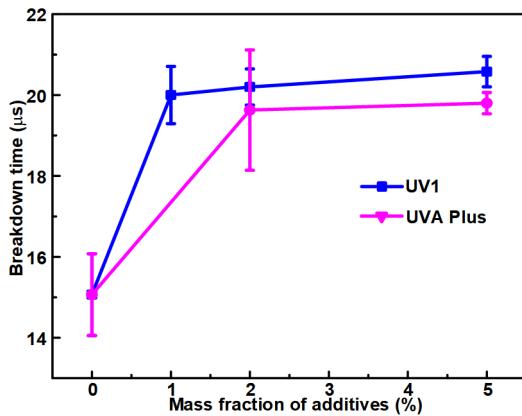
### A. BREAKDOWN CHARACTERISTICS

The results in Fig. 4a and Fig. 4b clearly indicated the increased  $V_{b-LI}$  and  $V_{a-LI}$  by the doping of UV1 and UVA Plus into NE oil. As the mass fraction of additives increases, both the  $V_{b-LI}$  and  $V_{a-LI}$  increases. Specifically,  $V_{a-LI}$  of the NE oil with UV1 increased rapidly. For example, 5 wt.% UV1 in the base liquid caused an increase in  $V_{b-LI}$  and  $V_{a-LI}$  of the base liquid by 15% (from 60.13 to 69.62 kV) and 60% (from 77.84 to 125.81 kV), respectively. Thus, the influence of UVA Plus on the lightning impulse dielectric properties of the NE oil was not significantly increased compared with UV1. Fig. 4 showed that 5 wt. % UVA plus in the base liquid caused an increase in the  $V_{b-LI}$  by 4% (from 60.13 to 62.96 kV) and  $V_{a-LI}$  by 17% (from 77.84 to 92.2 kV), respectively.

Fig. 5 showed the  $t_b$  of NE oil with doped molecules under  $V_{b-LI}$ . Briefly, the  $t_b$  of the modified NE oil initially elevated significantly with the growth of concentrations of doped molecules (UV1 and UVA Plus) and leveled off under certain mass fraction. Specifically, for the UV1, the  $t_b$  increased by 32.8% (from 15.06 to 20 us) with just 1 wt.% of doping. With additional 4 wt.% of UV1, the  $t_b$  of the modified NE oil increased by 3.2%. For the UVA Plus, for example, UV1 increased  $t_b$  by 36% and UVA Plus increased  $t_b$  only by 31.5% in the same mass fraction 5 wt.%. As the oil gap distances were the same, the average velocity of streamers in NE oil with UVA Plus under  $V_{b-LI}$  was more than that of NE oil with UV1 in the same mass fraction.

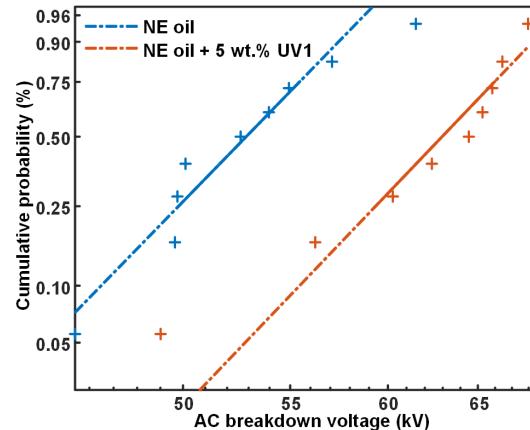


**FIGURE 4.** The positive LI acceleration voltage  $V_{a-LI}$  (a), the positive LI breakdown voltage  $V_{b-LI}$  (b) of NE oil with different mass fraction of additives.

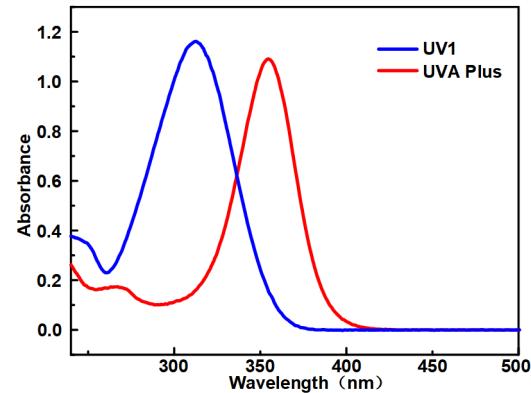


**FIGURE 5.** Positive LI breakdown time  $t_b$  under  $V_{b-LI}$  with different mass fraction of additives.

The AC breakdown voltage  $V_{b-AC}$  of NE oil with and without 5 wt.% UV1 were measured and the values were statistically analyzed with the Weibull distribution. The results were shown in Fig. 6. It is seen that 5 wt.% of UV1 in NE oil enhanced  $V_{b-AC}$  from 54.90 to 64.31 kV at the probability of 63.2%. The average  $V_{b-AC}$  increased from 52.78 to 61.99 kV, i.e. by 17% increased. It is a typically method to apply the shape parameter to evaluate the scattering of the measured  $V_{b-AC}$ , and higher corresponding to less scattering



**FIGURE 6.** The weibull plot of AC breakdown probability of NE oil.



**FIGURE 7.** UV-Vis absorption spectra of UV1 and UVA Plus.

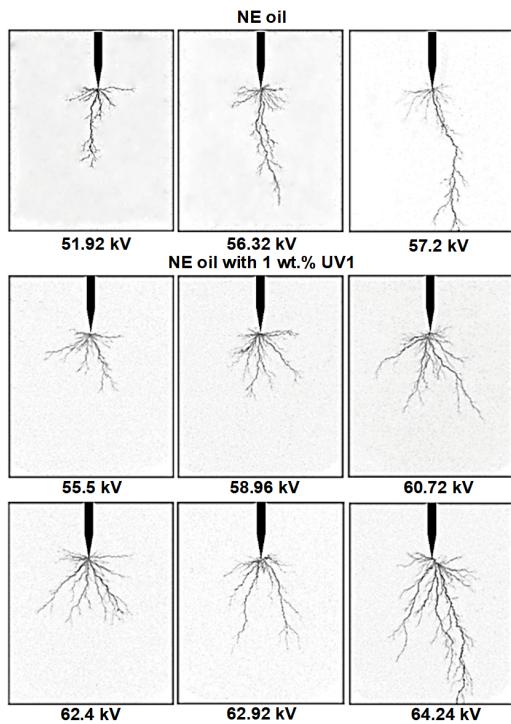
of the  $V_{b-AC}$  values. With 5 wt.% UV1 in NE oil, the  $\beta$  is found been raised from 12.04 to 16.13.

### B. UV-VIS ABSORPTION SPECTRA

The ultraviolet-visible (UV-Vis) absorption spectra of UV1 and UVA Plus was shown in Fig. 7. UV1 exhibited a higher absorption as compared to UVA Plus, which varied between 240 and 340 nm at a maximum absorption wavelength  $\lambda_{max}$  of 312 nm. Conversely, UVA Plus exhibited a higher absorption than UV1 between 340 and 410 nm at a maximum absorption wavelength  $\lambda_{max}$  of 355 nm. Therefore, during streamer propagation, UV1 is expected to have a better ability to absorb the higher energy ultraviolet irradiation and should benefit to restrict the ionization of the triglyceride molecules.

### C. STREAMER PROPAGATION BEHAVIOR

To better indicate the influence of UV1 on the streamer propagation behavior of the NE oil, Fig. 8 lists the shadowgraphic images of the streamers recorded by a high speed camera. It is clearly seen that the streamers in NE oil consisted of a number of luminous and slender branching filaments. Many small lateral branches were observed along the main filaments (1-2 pieces), but generally did not propagate lengthwise. As expected, the length of the streamer increased with increasing voltage. With the UV1 added in the NE oil, the



**FIGURE 8.** Shadowgraphic images of the streamers in the NE oil and with 1 wt.% UV1 added to the NE oil at different LI voltages.

filamentary lateral branches were able to develop further away from the needle tip and the number of main filaments (4-5 pieces) were much more than that of the pure NE oil. It was also quite visible that the streamer propagation in the NE oil with UV1 required a higher applied voltage to reach the same length as that in the pure NE oil. Another interesting point was that the streamer images captured at 64.24 kV exhibited a propagated head up to the plane electrode, while a voltage of 60 kV was sufficient to generate the breakdown in pure NE oil. As mentioned above, the UV1 increased the breakdown time from 15.06 to 20.58  $\mu$ s, which resulted in a decrease in the average velocity of streamer propagation from 1.66 to 1.21 km/s.

#### D. ELECTRONIC PROPERTY OF MOLECULES

The ionization potential (IP), electron affinity (EA), and first excitation energy (FE) of molecules in liquids are three molecular descriptors that qualitatively have been linked the streamer initiation and propagation processes [12], [18]. The results were shown in Table. 1. The calculated IP of UV1 (7.5 eV) and UVA Plus (7.4 eV) were almost identical and were quite close to the IP values of the triglyceride molecules (ca. 7.8 eV), which varied slightly due to the relatively similar molecular structures of the triglycerides [16]. Without the low IP molecules, the influence of the space charge shielding effect should not be considered with respect to the streamer propagation [19]. Furthermore, the IP of the triglycerides exhibited an inverse correlation with the applied electric field, which could increase the ionization rate of the triglyceride molecules under applied voltage [20]. The EA of

**TABLE 1.** The electronic property parameters of the molecules.

Molecule	IP (eV)	FE (eV)	EA (eV)
UVA Plus	7.4	2.7	0.49
UV1	7.5	2.9	0.25
Triolein	7.8	4.0	-0.35

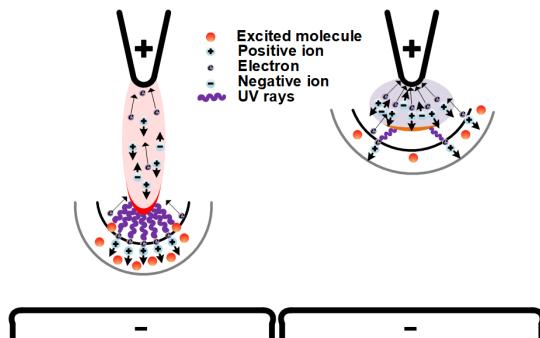
the molecules is another possible factor that may influence the streamer propagation. The EA of the UV1 and UVA Plus molecules were calculated as 0.25 and 0.49 eV, respectively. The EA of UV1 and UVA Plus were quite small, such that their influence on the propagation of the positive streamers may be deemed negligible [21]. The lower FE of the doping molecules in the ester liquids could inhibit the propagation of the streamer as compared to the base molecules [12]. The FE of UVA Plus and UV1 were lower than that of the triglyceride (4.0 eV). UVA Plus presented the FE value of 2.7 eV, which was around 0.2 eV lower than that of UV1. However, the effect of UVA Plus on the enhanced  $V_{a-LI}$  and  $V_{b-LI}$  of the NE oil was not as significant as that of UV1.

#### IV. DISCUSSIONS

The results obtained with UV1 demonstrate that there is probably an influence of ultraviolet absorption capacity of the doped molecules on the positive streamer propagation behaviors in the NE oils. Therefore, it is necessary to propose a hypothesis considering the restricted ionization dynamics in the streamer initiation and propagation of NE oils with molecules possessing absorption capacity in ultraviolet light.

When the applied voltage is beyond the streamer initial voltage, triglyceride molecules near the needle tip are ionized to form initial streamer branches [22], [23]. As the streamer initiation continues and tends to upgrade to propagation, the electric field from the space charges  $E_q$  dominates the streamer propagation. Charge carriers such as electrons and ions are accelerated by the electric field. Electrons play an important role in the collision process given their significantly higher mobility as compared to that of ions. When kinetic energy is less than the IP but more than the FE of triglyceride molecules, the electrons can lose its kinetic energy to excite triglyceride molecules during the collision process. The molecules in front of the streamer head can be excited by photons originating from the de-excitations of the excited molecules and these molecules can be ionized with less energy as compared to molecules in their ground state. The electric field in front of the streamer head increases to a level that allows the ionization of the surrounding triglyceride molecules, as shown in the left part of Fig. 9. The ionization of these molecules strengthens the electric field of the streamer head  $E$  as the feedback. This further increases  $E$  to enhance the electric field far from the needle tip [24]. The nonhomogeneous electric field distribution of the needle-plate electrode significantly enhances charge carrier transport at the axis of the needle-plate electrode system, as shown in the first row of Fig. 8.

With the UV1 in the NE oil, the streamer initiation also has many branches under LI voltage. When the streamer turns



**FIGURE 9.** Illustration for the effect of the ultraviolet absorbing molecules on the positive streamer propagation in the NE oils.

into propagation, the behavior of the streamer is different from that of the pure NE oil. According to the Planck-Einstein relation, the wavelength of the photon originating from the electron of the triglyceride molecule de-excites from the first excited state to the ground state:

$$\lambda = ch / (E_2 - E_1) = 310 \text{ nm} \quad (2)$$

where  $c$  is the velocity of light in the vacuum,  $h$  is the Planck constant,  $E_2$  is the electron energy of the higher energy level, and  $E_1$  is electron energy of the lower energy level.

The maximum absorption wavelength  $\lambda_{\max}$  of UV1 is 312 nm, which is similar to the wavelength of the light emissions from the de-excitations of the triglyceride molecules. The number of excited triglyceride molecules are decreased, and ionization rate is weakened during the development of the streamer in the NE oil, thereby resulting in lower space charges and therefore lower  $E_q$  values. Due to the geometry of the needle-plane electrode system and the lower contribution from  $E_q$ , the  $E$  is not high enough to allow significant ionization of the molecules at the axis of the electrode system farther away from needle tip. The streamer could only propagate at the position with relatively high applied electric field, i.e., not far away from the needle tip, as shown in the right part of Fig. 9. Therefore, the streamer branching increased and the length of the streamer in the NE oil with UV1 was shorter than that in the pure NE oil, which postponed the final arc formation and therefore improved the  $V_b\text{-LI}$  of NE oil.

The electric field strength at the electrode tip under  $V_a\text{-LI}$  was stronger than that under  $V_b\text{-LI}$ . The IP of the triglyceride presented an inverse correlation with the applied electric field, such that the ionization rate of the NE oil molecules under  $V_a\text{-LI}$  increased and thus enhanced  $E$ . When  $E$  reached a certain intensity, the IP of the triglyceride molecules could be less than or equal to their FE [20]. The ionization rate of the molecules sharply increased following photoionization and a large amount of space charge was generated during this propagation, which dramatically improved the velocity of the streamer in the NE oils [25]. As mentioned above, UV1 at a maximum absorption wavelength 312 nm exhibited energy (3.97 eV) that was similar to the FE of the triglyceride molecules. With the UV1 in NE oil, the photoionization should be inhibited during the streamer initiation

and propagation stages. This results in the decreased  $E$  and lacking of space charges, which are critical for the enhanced formation of the streamer (*cf.* Fig. 9). As a consequence, a higher field strength is necessary to enhance the  $E$ , which requires a sharp increase in the  $V_a\text{-LI}$  of the NE oil. The effect of UVA Plus at a maximum absorption wavelength of 354 nm presented good absorption to the long wavelength ultraviolet irradiation, such that increases in  $V_a\text{-LI}$  and  $V_b\text{-LI}$  were observed but were not as significant as that of UV1.

## V. CONCLUSION

In conclusion, we demonstrated that the LI and AC breakdown performance of the NE oil could be significantly improved by doping molecules with great absorption in ultraviolet lights. The maximum absorption wavelength of doped molecules should correspond to the first excitation energy of base liquid molecules. The ionization potential of doped molecules should be almost the same values as compared to the base molecule in the NE oils. The 5 wt.% of UV1 in NE oil has led to increases of positive  $V_b\text{-LI}$  and  $V_a\text{-LI}$  by 15% and 60%, respectively. The LI breakdown time was prolonged from 15.06 to 20.58  $\mu\text{s}$ , i.e. by 36% increased. The AC breakdown strength was noticed as 17% increased. However, the long wavelength absorption molecules UVA Plus (5 wt.% in NE oil) showed much less ability to enhance the positive  $V_b\text{-LI}$  and  $V_a\text{-LI}$  by 4.7% and 18.4%, respectively. The doped molecules in NE oils can decrease the number of excited triglyceride molecules and weaken ionization rate of molecules during the development of slow streamer. It can also inhibit the photoionization of the triglyceride molecules during the formation of fast streamer. The NE oil with such ultraviolet absorbing molecules exhibited good insulating properties and applicability in high voltage applications, in particular the formulation with UV1.

## REFERENCES

- [1] T. V. Oomen, "Vegetable oils for liquid-filled transformers," *IEEE Electr. Insul. Mag.*, vol. 18, no. 1, pp. 6–11, Jan. 2002.
- [2] C. P. McShane, K. J. Rapp, J. L. Corkran, G. A. Gauger, and J. Luksich, "Aging of Kraft paper in natural ester dielectric fluid," in *Proc. IEEE Int. Conf. Dielectr. Liquids*, Graz, Austria, Jul. 2012, pp. 173–177.
- [3] D. Martin, N. Lelekakis, W. Guo, and Y. Odarenko, "Further studies of a vegetable-oil-filled power transformer," *IEEE Electr. Insul. Mag.*, vol. 27, no. 5, pp. 6–13, Sep./Oct. 2011.
- [4] C. T. Duy, O. Lesaint, A. Denat, and N. Bonifaci, "Streamer propagation and breakdown in natural ester at high voltage," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 16, no. 6, pp. 1582–1594, Dec. 2009.
- [5] O. Lesaint and G. Massala, "Positive streamer propagation in large oil gaps: Experimental characterization of propagation modes," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 5, no. 3, pp. 360–370, Jun. 1998.
- [6] Q. Liu and Z. D. Wang, "Streamer characteristic and breakdown in synthetic and natural ester transformer liquids with pressboard interface under lightning impulse voltage," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 18, no. 6, pp. 1908–1917, Dec. 2011.
- [7] J. Hajek, J. Kranenborg, P. Sundqvist, R. Jonsson, T. Skytt, B. Samuelsson, R. Asano Jr, G. K. Frimppong and R. Girgis, "Considerations for the design manufacture and retro-filling of power transformers with high fire point biodegradable ester fluids," in *Proc. CIGRE General Session*, Paris, France, no. A2-203, 2012, pp. 1–9.
- [8] Y. Kamata and Y. Kako, "Flashover characteristics of extremely long gaps in transformer oil under non-uniform field conditions," *IEEE Trans. Electr. Insul.*, vol. EI-15, no. 1, pp. 18–26, Feb. 1980.

- [9] S. A. Ghani, N. A. Muhamad, Z. A. Noorden, H. Zainuddin, N. A. Bakar, and M. A. Talib, "Methods for improving the workability of natural ester insulating oils in power transformer applications: A review," *Electr. Pow. Syst. Res.*, vol. 163, pp. 655–667, Oct. 2018.
- [10] J. Li, Z. Zhang, P. Zou, S. Grzybowski, and M. Zahn, "Preparation of a vegetable oil-based nanofluid and investigation of its breakdown and dielectric properties," *IEEE Electr. Insul. Mag.*, vol. 28, no. 5, pp. 43–50, Sep. 2012.
- [11] Y. Zhong, Y. Lv, C. Li, Y. Du, M. Chen, S. Zhang, Y. Zhou, and L. Chen, "Insulating properties and charge characteristics of natural ester fluid modified by TiO<sub>2</sub> semiconductive nanoparticles," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 20, no. 1, pp. 135–140, Feb. 2013.
- [12] M. Unge, S. Singha, N. Van Dung, D. Linhjell, S. Ingebrigtsen, and L. E. Lundgaard, "Enhancements in the lightning impulse breakdown characteristics of natural ester dielectric liquids," *Appl. Phys. Lett.*, vol. 102, no. 17, pp. 172905–172909, Apr. 2013.
- [13] J. C. Devins, S. J. Rzad, and R. J. Schwabe, "Breakdown and prebreakdown phenomena in liquids," *J. Appl. Phys.*, vol. 52, no. 7, pp. 4531–4545, 1981.
- [14] *Methods for the Determination of the Lightning Impulse Breakdown Voltage of Insulating Liquids*, Standard IEC 60897, International Electrotechnical Commission, 1987.
- [15] A. D. Becke, "Density-functional thermochemistry. III. The role of exact exchange," *J. Chem. Phys.*, vol. 98, no. 7, pp. 5648–5652, Dec. 1992.
- [16] J. Li, Y. Wang, F. Wang, S. Liang, X. Lin, X. Chen, and J. Zhou, "A study on ionization potential and electron trap of vegetable insulating oil related to streamer inception and propagation," *Phys. Lett. A*, vol. 381, no. 44, pp. 3732–3738, Sep. 2017.
- [17] H. S. Smalø, Ø. Hestad, S. Ingebrigtsen, and P.-O. Åstrand, "Field dependence on the molecular ionization potential and excitation energies compared to conductivity models for insulation materials at high electric fields," *J. Appl. Phys.*, vol. 109, no. 12, pp. 073306–073320, Apr. 2011.
- [18] H. S. Smalø, P.-O. Åstrand, and S. Ingebrigtsen, "Calculation of ionization potentials and electron affinities for molecules relevant for streamer initiation and propagation," *IEEE Trans. Dielectr. Elecr. Insul.*, vol. 17, no. 3, pp. 733–741, Jun. 2010.
- [19] N. V. Dung, H. K. Høidalen, D. Linhjell, L. E. Lundgaard, and M. Unge, "Effects of reduced pressure and additives on streamers in white oil in long point-plane gap," *J. Phys. D, Appl. Phys.*, vol. 46, no. 25, pp. 255501–255516, Jun. 2013.
- [20] N. Davari, P.-O. Åstrand, S. Ingebrigtsen, and M. Unge, "Excitation energies and ionization potentials at high electric fields for molecules relevant for electrically insulating liquids," *J. Appl. Phys.*, vol. 113, no. 14, pp. 143707–143717, Mar. 2013.
- [21] S. Ingebrigtsen, "The influence of chemical composition on streamer initiation and propagation in dielectric liquids," Ph.D. dissertation, Fac. Natural Sci. Technol. Dept. Chem., Norwegian Univ. Sci. Technol., Trondheim, Norway, 2008.
- [22] D. André, "Conduction and breakdown initiation in dielectric liquids," in *Proc. IEEE Int. Conf. Dielectr. Liquids (ICDL)*, Trondheim, Norway, Jun. 2011, pp. 1–11.
- [23] W. Lu and Q. Liu, "Prebreakdown and breakdown mechanisms of an inhibited gas to liquid hydrocarbon transformer oil under positive lightning impulse voltage," *IEEE Trans. Dielectr. Elecr. Insul.*, vol. 23, no. 4, pp. 2450–2461, Aug. 2016.
- [24] J.-W. G. Hwang, "Elucidating the mechanisms behind pre-breakdown phenomena in transformer oil systems," Ph.D. dissertation, Massachusetts Inst. Technol., Cambridge, MA, USA, 2010.
- [25] L. Lundgaard, D. Linhjell, G. Berg, and S. Sigmond, "Propagation of positive and negative streamers in oil with and without pressboard interfaces," *IEEE Trans. Dielectrics Elecr. Insul.*, vol. 5, no. 3, pp. 388–395, Jun. 1998.



**SUNING LIANG** was born in Jiangsu, China, in 1991. He received the bachelor's degree from the Lanzhou University of Technology, Lanzhou, China, in 2014. He is currently pursuing the Ph.D. degree with the Electrical Engineering College, Chongqing University, Chongqing, China. His current research interests include partial discharge, pre-breakdown and breakdown phenomena in liquids, and natural ester dielectric liquids.



**FEIPENG WANG** (M'15) received the Ph.D. degree in materials physics and chemistry from Tongji University, China, in 2007. He was with the Applied Condensed-Matter Physics Group, University of Potsdam, and with the Fraunhofer Institute for Applied Polymer Research, Germany, from 2007 to 2013, with a focus on functional dielectrics and their applications. Since 2014, he has been a Professor with Chongqing University, China, with an emphasis on engineering dielectrics and applications in power grid.



**ZHENGYONG HUANG** received the Ph.D. degree in electrical engineering from Chongqing University, China, in 2016, where he is currently a Lecturer. His current research interests include the development of natural ester-based ecofriendly nanofluid, bio-inspired nano-coating and structured nanocomposite for dielectric, and electrical insulation.



**WEIGEN CHEN** was born in Zhejiang, China, in 1967. He received the B.Sc., M.Sc., and Ph.D. degrees in electrical engineering from Chongqing University, China, in 1990, 1993, and 2003, respectively, where he is currently a Professor with the Department of Electrical Engineering. He has published two books and over 130 papers in his professional work. His current research interests include online monitoring and fault diagnosis of power equipment, condition-based maintenance, and the internal insulation and thermal properties of power transformers. He is a member of the CIGRÉ WG A2.27 working groups.



**YOUYUAN WANG** received the M.S. and Ph.D. degrees in electrical engineering from Chongqing University, Chongqing, China, in 2003 and 2008, respectively, where he is currently a Professor with the School of Electrical Engineering. He has authored or coauthored more than 50 journal papers and 30 papers published in the proceedings of international conferences. His current research interests include dielectric properties and space charge behavior of nanocomposites, online detection of insulation condition in electrical devices, and insulation fault diagnosis for high-voltage equipment.



**JIAN LI** (M'05–SM'11) received the M.S. and Ph.D. degrees in electrical engineering from Chongqing University, Chongqing, China, in 1997 and 2001, respectively, where he is currently a Professor and the Dean of the School of Electrical Engineering. He has authored or coauthored more than 130 journal papers and 90 papers published in the proceedings of international conferences. His current research interests include online condition monitoring and fault diagnosis of HV equipment, environment-friendly insulation materials, and nano dielectrics.