IEEE Access
Multidisciplinary : Rapid Review : Open Access Journal

Received May 22, 2020, accepted June 13, 2020, date of publication June 17, 2020, date of current version June 29, 2020.

Digital Object Identifier 10.1109/ACCESS.2020.3003246

Statistical Investigation of Lightning Impulse Breakdown Voltage of Natural and Synthetic Ester Oils-Based Fe_3O_4, Al_2O_3 and SiO_2 Nanofluids

ABDERRAHMANE BEROUAL[1], (Fellow, IEEE), AND USAMA KHALED[2,3]
[1]AMPERE Laboratory, CNRS UMR 5005, École Centrale de Lyon, University of Lyon, 69134 Ecully, France
[2]Department of Electrical Engineering, Faculty of Energy Engineering, Aswan University, Aswan 81528, Egypt
[3]Department of Electrical Engineering, College of Engineering, King Saud University, Riyadh 11421, Saudi Arabia

Corresponding author: Usama Khaled (usamakhaled@energy.aswu.edu.eg)

ABSTRACT The characteristics of lighting impulse breakdown voltage (LI-BDV) constitute key performance indicators of insulating materials, especially in insulations based on liquids and liquid-paper for high voltage applications. Encouraged by environmental policies worldwide, new insulating liquids have been developed in recent years. Among these are natural and synthetic esters that are currently considered as potential substitutes for traditional mineral oils due to their environmentally friendly properties. Recent technological advances have shown that the addition of specific nanoparticles (NPs) to insulating liquids can improve both the dielectric withstand voltage and the thermal behavior of liquid insulations. This paper examines the effects of some NPs on the negative LI-BDV of natural and synthetic ester liquids, namely MIDEL 1204 and MIDEL 7131, respectively. The used NPs are Fe_3O_4, Al_2O_3 and SiO_2. The LI-BDV measurements are executed in accordance with IEC 60060 standard, and the breakdown voltage with 1% probability risk (i.e., the lowest possible breakdown voltage) was also estimated. The experimental results showed that these NPs improved the LI-BDV of both synthetic and natural ester liquids and an optimal concentration of NPs can be determined. Statistical analysis was also performed to check the compliance of the experimental results with the most common probability distributions, normal and Weibull distributions.

INDEX TERMS $U_{50\%}$ breakdown voltage, nanofluids, natural ester, synthetic ester, statistical analysis, Weibull distribution, normal distribution.

I. INTRODUCTION

Natural and synthetic esters are increasingly used in oil-filled apparatus, especially in high voltage (HV) transformers, to replace the commonly used mineral oil (MO). As well as some of their interesting properties from an environmental point of view, such as biodegradability and non-toxicity [1]–[4], these insulating liquids have a high flash point and low sensitivity to water content. These characteristics qualify them for use in devices installed near urban areas. However, the dielectric withstand of these esters is less good than that of MO [5]. In addition, their viscosity is 3 to 4 times higher than that of MO, which constitutes a disadvantage for heat transfer and thus for the cooling of power transformers. To solve this problem, one of the solutions would be the addition of specific nanoparticles (NPs) to the esters. Indeed, the addition of some NPs considerably improve the thermal properties of liquids; nanofluids (NFs) are commonly used for cooling of various devices and systems [6]–[8]. However, according to recent studies, the addition of some NPs also enhances the AC breakdown voltage (AC-BDV) of insulating liquids and especially that of MO-based Fe_3O_4 nanofluids [9]. In the case of esters, the AC-BDV can also be improved, but to a lesser degree than that of MO [10], [11].

Nanofluids thus appear to be potential substitutes for MO, since they fulfill two of the essential properties for use in HV devices and in particular in power transformers, namely efficient heat transfer and good dielectric performances.

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

Note that, due to the oxidation concern, the use of natural esters is limited to sealed oil-filled apparatus such as sealed rectifier transformers, tap-changers and power transformers up to 550 kV as those produced by General Electric [12]. Large sealed transformers are rare and very expensive. However, despite this limitation for natural esters, the higher permissible moisture content of both natural and synthetic esters makes the moisture resulting from thermal aging of paper insulation readily absorbed by ester liquids, which slows the paper aging process by keeping the paper drier and helps extend the life of the transformer. In addition, ester liquids have higher electrical permittivity than mineral oils which is an advantage in terms of electrical field distribution in insulating system of a power transformer.

Oil-filled devices and their components, including the insulating oils, must also withstand certain constraints including lightning over-voltages. These constitute key performance indicators of insulating materials, in particular in insulations based on liquids and liquid-paper for HV applications.

To this end, some investigations have taken place into the LI-BDV of NFs [13]–[20]. It has been reported that, in a divergent electric field (point-plane electrode configuration); Fe_3O_4 NPs significantly increased the positive LI-BDV of MO [13]. The same results were found for the negative LI-BDV; however, the improvement is less important than for the positive polarity [13], [14]. The polarity effect will not be observed in uniform or quasi-uniform electric fields because of the symmetry of electrode configuration.

The aim of this work is to study the LI-BDV of natural ester (NE) and synthetic ester (SE) liquid-based NFs in a sphere-to-sphere electrode configuration (i.e., in a quasi-uniform electric field), using the same NPs as in previous work in AC [10], [11]. The main objective of this study is to compare the dielectric withstand in lightning impulse voltages to those measured in AC in our previous work, under the same experimental conditions and for the same nanofluids. Weibull and normal statistical analyses are performed to verify the compliance of the experimental findings with one or both of these probabilistic laws. The interest of such study is, among other things, the prediction of failures of nanofluids-filled apparatus in the event of lightning overvoltages and therefore the protection of HV equipment.

II. EXPERIMENTAL TECHNIQUE

MIDEL 7131 SE and MIDEL 1204 NE are chosen as the base liquids in this study; their main characteristic parameters are given in Table 1. The NPs used for preparing nanofluid samples are (1) insulating NPs of Al_2O_3 and SiO_2 of 50 nm size; and (2) magnetic NPs of Fe_3O_4 of 50 nm size. These were supplied by Sigma-Aldrich. Their distribution, composition, shape and size as well as the techniques used for their characterization were described in previous works [9], [10].

The breakdown voltage (BDV) tests were executed under negative lightning impulse, on a test cell of 500 ml volume containing two brass spherical electrodes of 12.5 mm diameter with the electrode gap being 2.50 ± 0.05 mm. All measurements were performed under continuous stirring. The lightning impulse (LI) voltage was supplied by a 1 MV - 40 kJ - 1.2/50 μs Marx generator (Haefely type) connected to a dedicated capacitive divider; note that only three stages of the generator were used for a maximum voltage of 300 kV. The $U_{50\%}$ breakdown voltage is determined by the up-and-down method according to IEC 60060 ed3 2010-09.

TABLE 1. Physicochemical properties of synthetic MIDEL 7131 and Natural MIDEL 1204 Ester Liquids.

Property	MIDEL 7131	MIDEL 1204
Density at 20 °C (kg/dm^3)	0.97	0.92
Kinematic viscosity at 40 °C (cSt)	29	37
Pour point (°C)	−56	−31
Flash point (°C)	260	> 315
Fire point (°C)	316	> 350
Total acid number (mg KOH/g)	< 0.03	> 0.04
Water content (ppm)	300	100
Dissipation factor at 90 °C	0.8 %	0.9 %

The method involves performing first some preliminary tests to determine the proper voltage from which one starts the series of measurements. Once this voltage has been determined, a series of 24 measurements is started by increasing by 3 kV if there is no breakdown or by decreasing by 3 kV in the event of breakdown and so on; 24 readings are taken to be sufficient for statistical analysis of Weibull and normal distributions.

Note that most researchers use the step-by-step method following IEC 60897 standard. However, some studies also used the up-and-down method [21].

III. EXPERIMENTAL RESULTS: STATISTICAL ANALYSIS

This section presents the results of $U_{50\%}$ LI-BDV measurements of NFs' samples and their compliance with the normal and Weibull distribution laws [22], [23]. These probabilistic distributions are the most used to analyze the BDV of dielectrics. The interest in such analyses for the design and maintenance of power apparatus as well as for improving the predictability of insulation performance has been outlined in previous work [10]. Note that in this study one uses the 2-parameter Weibull distribution assuming the location parameter equal to 0 instead 3-parameter Weibull distribution.

Two hypothesis tests are applied to verify to which probabilistic law the experimental data complies, namely the Shapiro–Wilk test [23] and the Anderson–Darling test [24]. The details of the methods of application of these tests and computation of the related parameters have been described elsewhere [25]. The significance level α to which the p-values are compared in order to decide whether or not the experimental results comply with the statistical distribution is taken to be 5%. Recall that for the p-values less than or equal to α, the null hypothesis is rejected and, consequently, the data of the sample do not belong to the law of statistical distribution considered. Smaller p-values provide stronger evidence against the null hypothesis. Larger values for the Anderson-Darling statistic indicate that the data do not follow a Weibull distribution.

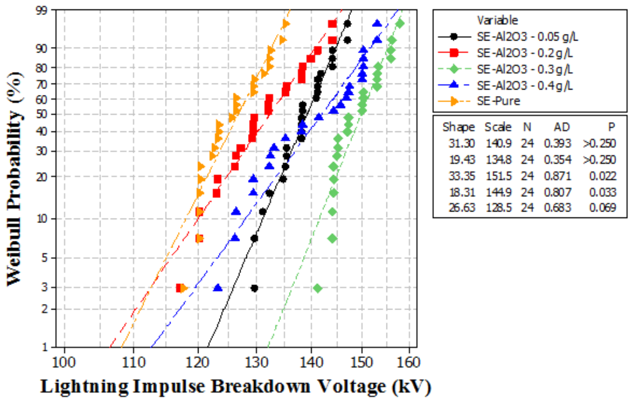

FIGURE 1. Weibull distribution of the LI breakdown voltages of SE/Al_2O_3 nanofluid.

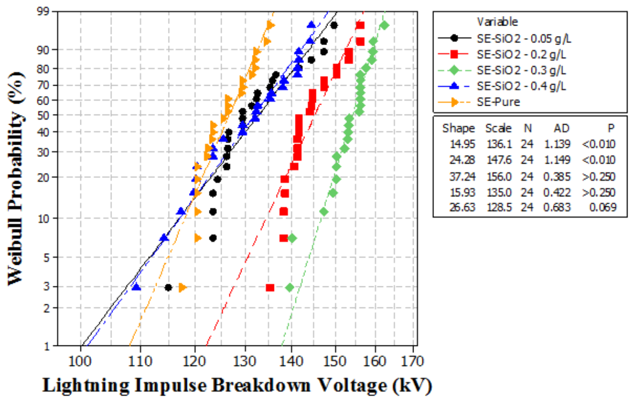

FIGURE 2. Weibull distribution of the LI breakdown voltages of SE/SiO_2 nanofluid.

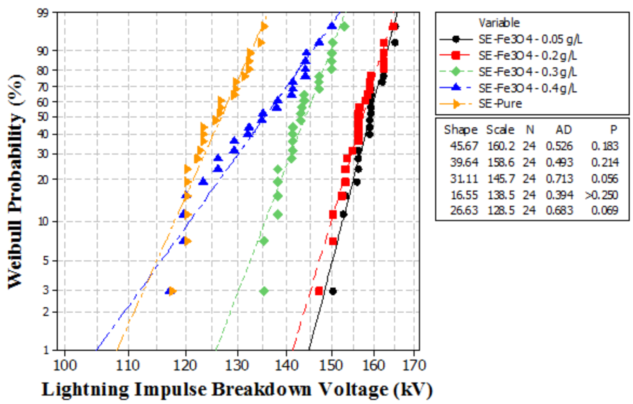

FIGURE 3. Weibull distribution of the LI breakdown voltages of SE/Fe_3O_4 nanofluid.

A. WEIBULL DISTRIBUTION

1) SYNTHETIC ESTER LIQUID – BASED NANOFLUIDS

Figures 1 to 3 depict the Weibull LI-BDV plots of SE-based Fe_3O_4, Al_2O_3 and SiO_2 NFs with different concentrations of NPs. Beside each figure are given the main parameters characterizing the Weibull distribution, namely the shape parameter representing the slope of the line, the scale parameter related to the scattering of the data and indicating the degree of failure, the Anderson–Darling (AD) and the *p*-values.

TABLE 2. Hypothesis test of compliance with Weibull distribution of the average LI-BDV of SE-based nanofluids.

Sample	*p*-value	Conformity to Weibull distribution
Pure SE	0.069	Accepted
SE/Fe_3O_4 (0.05 g/L)	0.183	Accepted
SE/Fe_3O_4 (0.2 g/L)	0.214	Accepted
SE/Fe_3O_4 (0.3 g/L)	0.056	Accepted
SE/Fe_3O_4 (0.4 g/L)	>0.250	Accepted
SE/Al_2O_3 (0.05 g/L)	>0.250	Accepted
SE/Al_2O_3 (0.2 g/L)	>0.250	Accepted
SE/Al_2O_3 (0.3 g/L)	0.022	Rejected
SE/Al_2O_3 (0.4 g/L)	0.033	Rejected
SE/ SiO_2 (0.05 g/L)	<0.010	Rejected
SE/ SiO_2 (0.2 g/L)	<0.010	Rejected
SE/ SiO_2 (0.3 g/L)	>0.250	Accepted
SE/ SiO_2 (0.4 g/L)	>0.250	Accepted

TABLE 3. $U_{50\%}$ LI-BDV of SE-Based Nanofluids.

	SE	Fe_3O_4	Al_2O_3	SiO_2
	SE / 0.05 (g/L) NF			
BDV (kV)	126.06	158.29	138.55	131.89
Increment (%)		25.57	9.91	4.62
	SE / 0.2 (g/L) NF			
BDV (kV)	126.06	156.53	131.32	144.63
Increment (%)		24.17	4.17	14.73
	SE / 0.3 (g/L) NF			
BDV (kV)	126.06	143.35	149.15	153.69
Increment (%)		13.72	18.32	21.84
	SE/ 0.4 (g/L) NF			
BDV (kV)	126.06	134.09	140.58	130.47
Increment (%)		6.37	11.52	3.50

The *p*-values and compliance of experimental data with the Weibull distribution are summarized in Table 2. It appears that the experimental results generally comply with this probabilistic distribution law, apart from SE with Al_2O_3 at concentrations of 0.3 g/L and 0.4 g/L, and SE with SiO_2 at concentrations of 0.05 g/L and 0.2 g/L.

Table 3 summarizes the $U_{50\%}$ values of LI-BDV. It is noted that the tested NPs improve the LI-BDV of SE. The best improvements are approximately 25.6% with Fe_3O_4 at a concentration of 0.05 g/L; 18.3% with Al_2O_3 at a concentration of 0.3 g/L; and 22% of SiO_2 at a concentration of 0.3 g/L. It is also noticed from Table 3, the existence of an optimal concentration for each type of NPs, in the investigated concentrations range.

Among the interesting and useful parameters from a practical viewpoint, in particular for the design of insulating components, is the LI-BDV at 1% cumulative probability, this being the lowest possible BDV. Table 4 summarizes the LI-BDV at 1% probability for investigated NFs. It can be observed that for each type of NP, the 1% values of LI-BDV increase up to an optimal concentration corresponding to the highest value and then decrease.

Note that the $U_{50\%}$ value of LI-BDV of pure SE of 126.06 kV is more than twice the mean value of 60 kV of the BDV measured under AC-BDV.

TABLE 4. LI-BDV at 1% cumulative probability of SE-based nanofluids (from weibull distribution).

Breakdown voltage probability (%)	Breakdown voltage (kV)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Increment (%)
	SE	0.05 g/L		0.2 g/L		0.3 g/L		0.4 g/L	
	Fe_3O_4 Nanoparticles								
	108.1	144.8	34.0	141.2	30.6	125.7	16.3	104.9	-3.0
1%	Al_2O_3 Nanoparticles								
	108.1	121.6	12.5	106.4	-1.6	132.0	22.1	112.7	4.3
	SiO_2 Nanoparticles								
	108.1	100.1	-7.4	122.1	13.0	137.8	27.5	101.1	-6.5

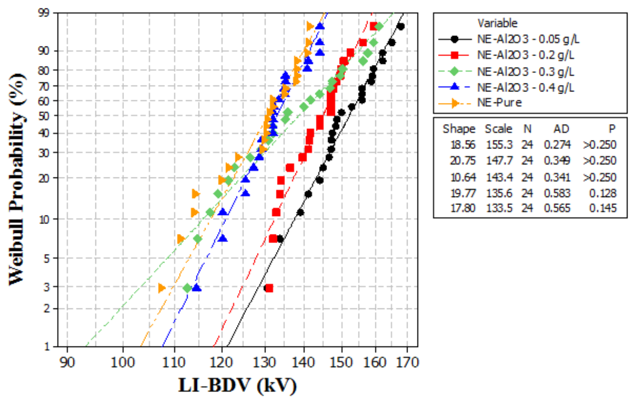

FIGURE 4. Weibull distribution of the LI breakdown voltages of NE/Al_2O_3 nanofluid.

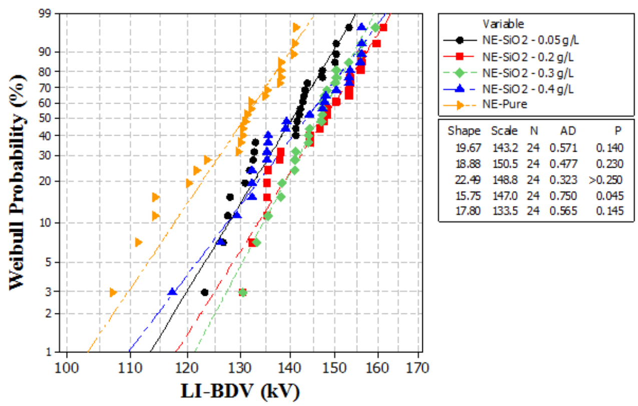

FIGURE 5. Weibull distribution of the LI breakdown voltages of NE/SiO_2 nanofluid.

2) NATURAL ESTER LIQUID – BASED NANOFLUIDS

Figures 4 to 6 show the Weibull plots of the LI-BDV of NE-based Fe_3O_4, Al_2O_3 and SiO_2 nanofluids. The characteristic parameters of these curves are indicated beside each figure. Table 5 summarizes the *p*-values and compliance of experimental data with the Weibull distribution for investigated NFs. It should be noted that the experimental data generally follow the Weibull distribution with the exception of three mixtures, namely NE/Fe_3O_4 (0.05 g/L), NE/Fe_3O_4 (0.4 g/L) and NE/ SiO_2 (0.4 g/L) for which the *p*-values are lower than 5% that is the level of significance chosen for the tests.

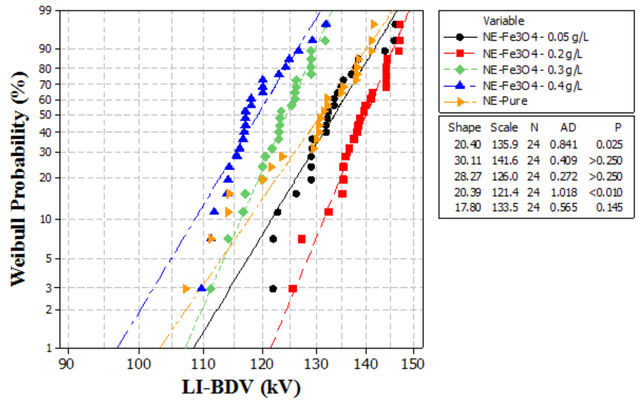

FIGURE 6. Weibull distribution of the LI breakdown voltages of NE/Fe_3O_4 nanofluid.

TABLE 5. Hypothesis test of compliance with weibull distribution of the average LI-BDV of NE-based nanofluids.

Sample	*p*-value	Conformity to Weibull distribution
Pure NE	0.145	Accepted
NE/Fe_3O_4 (0.05 g/L)	0.025	Rejected
NE/Fe_3O_4 (0.2 g/L)	>0.250	Accepted
NE/Fe_3O_4 (0.3 g/L)	>0.250	Accepted
NE/Fe_3O_4 (0.4 g/L)	<0.010	Rejected
NE/Al_2O_3 (0.05 g/L)	>0.250	Accepted
NE/Al_2O_3 (0.2 g/L)	>0.250	Accepted
NE/Al_2O_3 (0.3 g/L)	>0.250	Accepted
NE/Al_2O_3 (0.4 g/L)	0.128	Accepted
NE/ SiO_2 (0.05 g/L)	0.140	Accepted
NE/ SiO_2 (0.2 g/L)	0.230	Accepted
NE/ SiO_2 (0.3 g/L)	>0.250	Accepted
NE/ SiO_2 (0.4 g/L)	0.045	Rejected

It should first be noted that $U_{50\%}$ of the LI-BDV value of pure NE is also almost twice that of the mean value of the AC-BDV (i.e., 129.34 kV against 68.77 kV). However, it is observed that the NPs under consideration improve the LI-BDV of NE (Table 6); and for each type of NPs, there exists, in the range of the studied concentrations, an optimal concentration giving the highest value of $U_{50\%}$ LI-BDV. The best improvements are approximately 7.5% with Fe_3O_4 at a concentration of 0.2 g/L; 16.8% with Al_2O_3 at a concentration of 0.05 g/L; and 13% of SiO_2 at a concentration of 0.2 g/L.

The analysis of the LI-BDV at 1% cumulative probability also shows that NPs enhance the LI-BDV. For each type of NPs, the variation in 1% values of LI-BDV with the concentration of NPs displays an optimal concentration corresponding to the highest value and then decreases (Table 7). The best improvement is approximately 17.6% for the three types of NPs, but at different concentrations of NPs: 0.2 g/L with Fe_3O_4, 0.05 g/L with Al_2O_3 and 0.3 g/L with SiO_2.

TABLE 6. $U_{50\%}$ **LI-BDV of natural ester-Based nanofluids.**

	NE	Fe_3O_4	Al_2O_3	SiO_2
	NE / 0.05 (g/L) NF			
BDV (kV)	129.34	132.73	151.02	139.37
Increment (%)		2.62	16.76	7.75
	NE / 0.2 (g/L) NF			
BDV (kV)	129.34	139.05	144.08	146.28
Increment (%)		7.51	11.40	13.10
	NE / 0.3 (g/L) NF			
BDV (kV)	129.34	123.59	136.90	145.43
Increment (%)		-4.45	5.85	12.44
	NE/ 0.4 (g/L) NF			
BDV (kV)	129.34	118.62	132.08	142.09
Increment (%)		-8.29	2.12	9.86

TABLE 7. **LI-BDV at 1% cumulative probability of NE-based nanofluids (from weibull distribution).**

Breakdown voltage probability (%)	Breakdown voltage (kV)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Increment (%)
	NE	**0.05 g/L**		**0.2 g/L**		**0.3 g/L**		**0.4 g/L**	
				Fe_3O_4 Nanoparticles					
	103.1	108.5	5.2	121.6	17.9	107.1	3.9	96.9	-6.0
1%				**Al_2O_3 Nanoparticles**					
	103.1	121.2	17.6	118.3	14.7	93.1	-9.7	107.4	4.2
				SiO_2 Nanoparticles					
	103.1	113.4	10.0	117.9	14.4	121.2	17.6	109.8	6.5

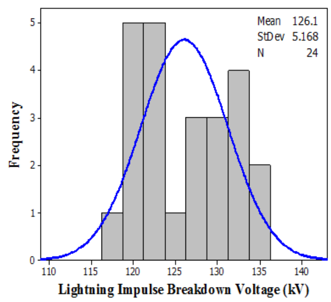

FIGURE 7. **LI Histogram of pure SE.**

B. NORMAL DISTRIBUTION

1) SYNTHETIC ESTER LIQUID – BASED NANOFLUIDS

The histograms of the distribution of the $U_{50\%}$ LI-BDV values of the tested samples and the corresponding skewness and kurtosis are given in Figures 7 to 10. This representation enables to detect possible anomalies in the distribution of the BDV in different ranges of voltages. So, it appears from the computed p-values that the experimental findings do not completely comply with the normal distribution. Indeed, the hypothesis tests show that the p-values for SE/Al_2O_3 (0.4 g/L), SE/SiO_2 (0.2 g/L) and SE/SiO_2 (0.3 g/L) NF samples are lower than 5% as shown on Table 8.

Regarding the skewness and kurtosis values, they indicate that the SE-based NFs results do not rigorously comply with the normal distribution since they are different from 0 and 3, respectively; the coefficients of skewness and kurtosis of the normal distribution have to be respectively 0 and 3. The experimental findings deviate somewhat from the normal distribution since the skewness values vary between −1.2 and +0.6. In addition, the highest value of the kurtosis is less than 1.4; thus the distribution is slightly

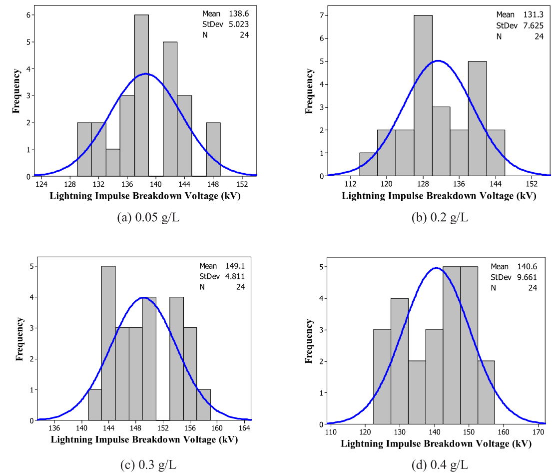

FIGURE 8. **a. Histogram of LI SE/Al_2O_3. b. Skewness and kurtosis of LI SE/Al_2O_3.**

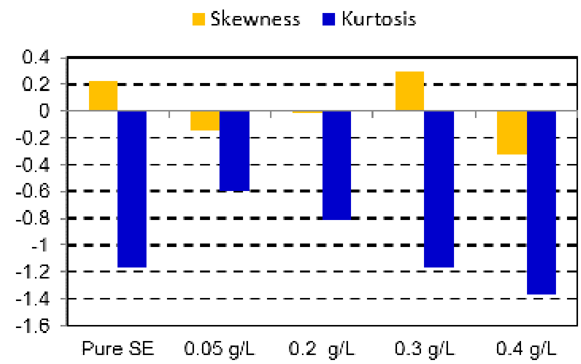

FIGURE 8. ***(Continued)*** **a. Histogram of LI SE/Al_2O_3. b. Skewness and kurtosis of LI SE/Al_2O_3.**

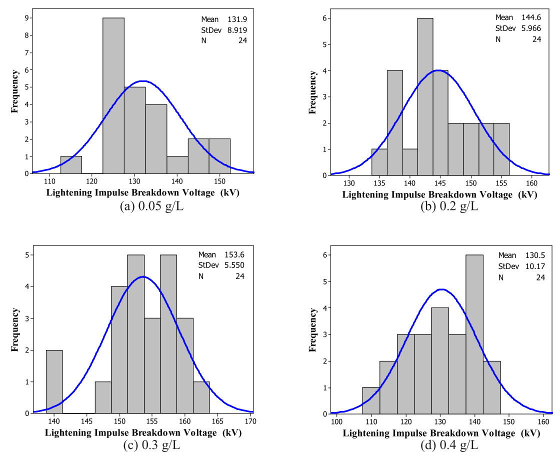

FIGURE 9. **a. Histogram of LI SE/SiO_2. b. Skewness and kurtosis of LI SE/SiO_2.**

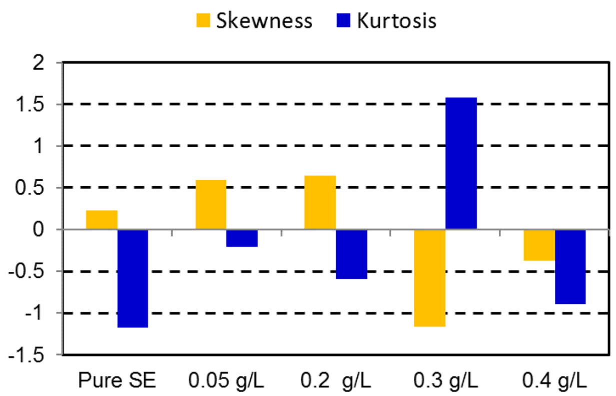

FIGURE 9. *(Continued)* **a. Histogram of LI SE/SiO_2. b. Skewness and kurtosis of LI SE/SiO_2.**

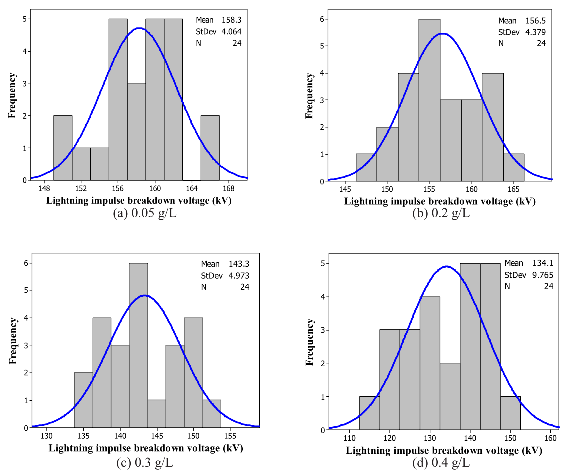

FIGURE 10. **a. Histogram of LI SE/Fe_3O_4. b. Skewness and kurtosis of LI SE/Fe_3O_4.**

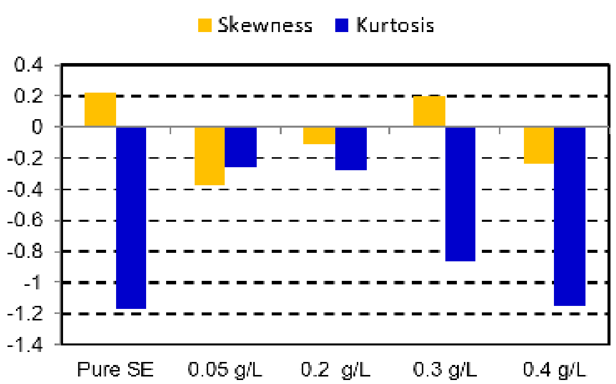

FIGURE 10. *(Continued)* **a. Histogram of LI SE/Fe_3O_4. b. Skewness and kurtosis of LI SE/Fe_3O_4.**

platykurtic [25]. Note that the skewness and kurtosis values are computed using the statistical package - *Statistical Package for the Social Sciences* (SPSS) [26].

2) NATURAL ESTER LIQUID – BASED NANOFLUIDS

Figures 11 to 14 depict the histograms of the distribution of BDV of the tested samples and the corresponding skewness and kurtosis. By applying the hypothesis test and by computing the *p*-values, it is noticed that all experimental data comply with the normal distribution except pure NE, as shown on Table 9.

Note, even if the *p*-values indicated that the distribution of experimental results of all NFs obeys that normal law, the skewness values are not null and the kurtosis scores vary between −0.8 and + 0.8. This indicates that the distribution of experimental results slightly deviates from the normal law.

TABLE 8. **Hypothesis test of compliance with normal distribution of the average LI-BDV of SE-based nanofluids.**

Sample	*p*-value	Conformity to normal distribution
Pure SE	0.151	Accepted
SE/Fe_3O_4 (0.05 g/L)	0.149	Accepted
SE/Fe_3O_4 (0.2 g/L)	0.563	Accepted
SE/Fe_3O_4 (0.3 g/L)	0.234	Accepted
SE/Fe_3O_4 (0.4 g/L)	0.234	Accepted
SE/Al_2O_3 (0.05 g/L)	0.513	Accepted
SE/Al_2O_3 (0.2 g/L)	0.831	Accepted
SE/Al_2O_3 (0.3 g/L)	0.054	Accepted
SE/Al_2O_3 (0.4 g/L)	0.032	Rejected
SE/ SiO_2 (0.05 g/L)	0.051	Accepted
SE/ SiO_2 (0.2 g/L)	0.039	Rejected
SE/ SiO_2 (0.3 g/L)	0.024	Rejected
SE/ SiO_2 (0.4 g/L)	0.285	Accepted

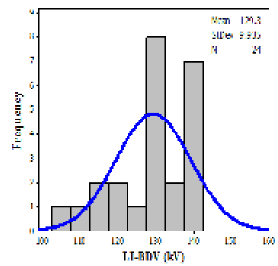

FIGURE 11. **LI Histogram of pure NE.**

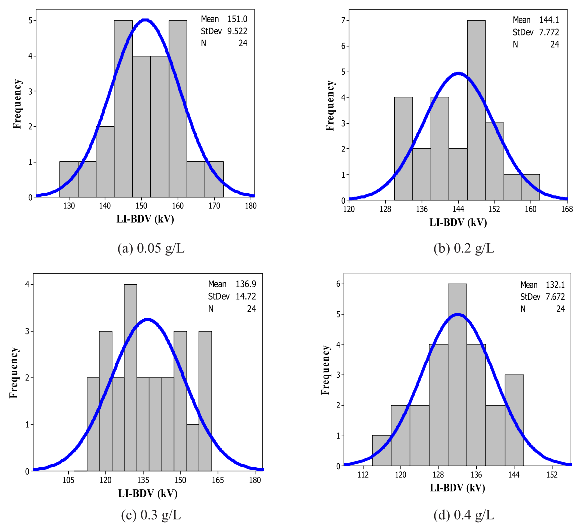

FIGURE 12. **a. Histogram of LI NE/Al_2O_3. b. Skewness and kurtosis of LI NE/Al_2O_3.**

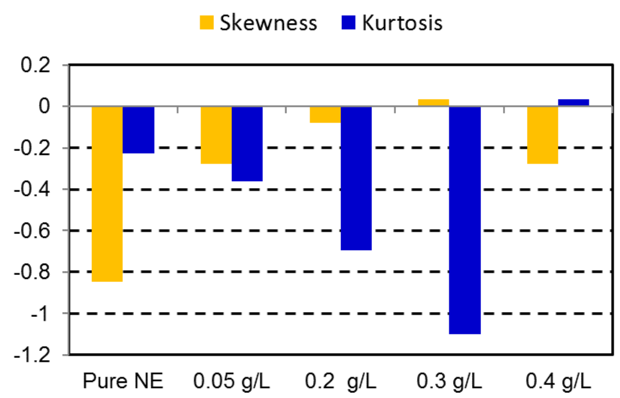

FIGURE 12. ***(Continued)*** **a. Histogram of LI NE/Al_2O_3. b. Skewness and kurtosis of LI NE/Al_2O_3.**

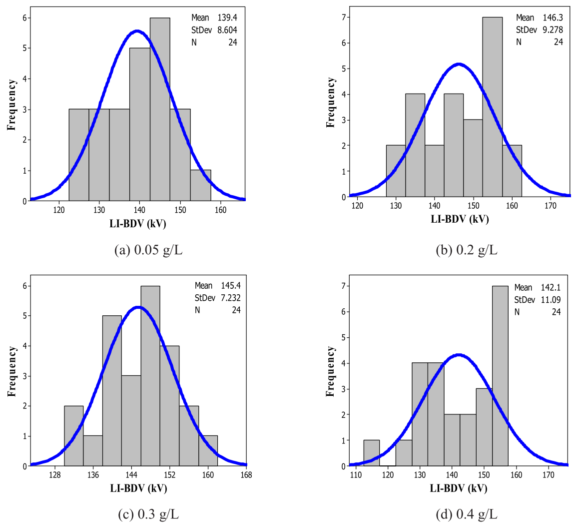

FIGURE 13. **a. Histogram of LI NE/SiO_2. b. Skewness and kurtosis of LI NE/SiO_2.**

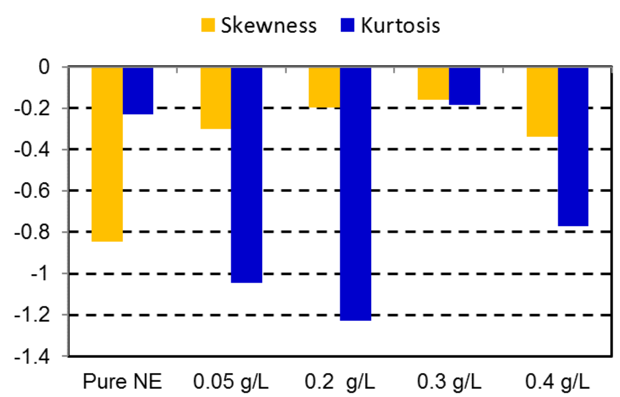

FIGURE 13. ***(Continued)*** **a. Histogram of LI NE/SiO_2. b. Skewness and kurtosis of LI NE/SiO_2.**

Note that most of the standard deviations are less than 7%, except NE/Al_2O_3 at a concentration of 0.3 g/L in which case it is of about 10.8%.

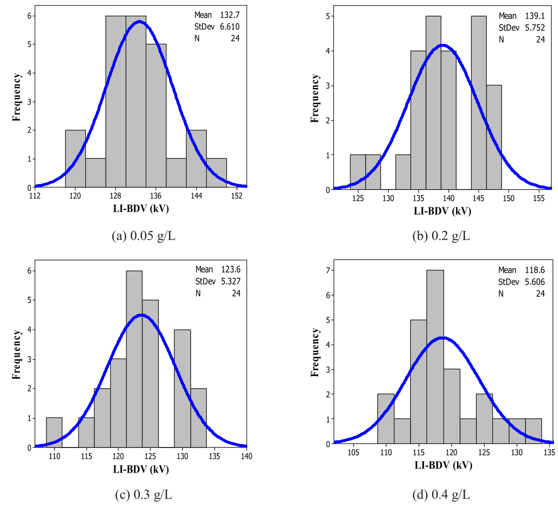

FIGURE 14. **a. Histogram of LI NE/Fe_3O_4. b. Skewness and kurtosis of LI NE/Fe_3O_4.**

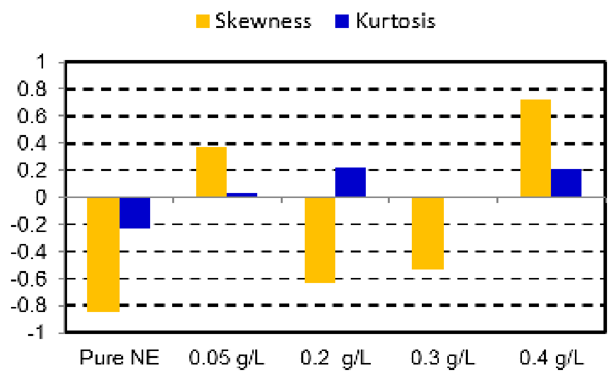

FIGURE 14. ***(Continued)*** **a. Histogram of LI NE/Fe_3O_4. b. Skewness and kurtosis of LI NE/Fe_3O_4.**

TABLE 9. **Hypothesis test of compliance with normal distribution of the average LI-BDV of NE-based nanofluids.**

Sample	*p*-value	Conformity to normal distribution
Pure NE	0.020	Rejected
NE/Fe_3O_4 (0.05 g/L)	0.333	Accepted
NE/Fe_3O_4 (0.2 g/L)	0.294	Accepted
NE/Fe_3O_4 (0.3 g/L)	0.502	Accepted
NE/Fe_3O_4 (0.4 g/L)	0.182	Accepted
NE/Al_2O_3 (0.05 g/L)	0.763	Accepted
NE/Al_2O_3 (0.2 g/L)	0.330	Accepted
NE/Al_2O_3 (0.3 g/L)	0.730	Accepted
NE/Al_2O_3 (0.4 g/L)	0.334	Accepted
NE/ SiO_2 (0.05 g/L)	0.062	Accepted
NE/ SiO_2 (0.2 g/L)	0.119	Accepted
NE/ SiO_2 (0.3 g/L)	0.816	Accepted
NE/ SiO_2 (0.4 g/L)	0.091	Accepted

IV. DISCUSSION

Based on the aforedescribed results,

1) The $U_{50\%}$ value of LI-BDV of pure SE is more than twice that of the mean value of BDV measured under

AC-BDV; (i.e., 126.06 kV against 60 kV) [11]. Similar results are found with NE, where the $U_{50\%}$ of the LI-BDV value of pure NE is also almost twice that of the mean value of the AC-BDV (i.e., 129.34 kV against 68.77 kV) [10].

2) The LI-BDVs of both natural and synthetic esters are enhanced when adding NPs. Note that for each type of NP, there is an optimal concentration that gives the highest value of $U_{50\%}$ LI-BDV for both ester liquids.

For SE, the best improvement of the $U_{50\%}$ value of LI-BDV is obtained with magnetic NPs (Fe_3O_4), as is also the case of the AC-BDV [11]. For the LI-BDV, the value is increased by approximately 25.6% with Fe_3O_4 at a concentration of 0.05 g/L; 22% with SiO_2 at a concentration of 0.3 g/L; and 18.3% with Al_2O_3 at a concentration of 0.3 g/L. The improvement of the AC-BDV is approximately 48% with Fe_3O_4, 32% with SiO_2 and 25% with Al_2O_3; respectively.

For NE, the best improvements of the $U_{50\%}$ value of LI-BDV are approximately 7.5% with Fe_3O_4 at a concentration of 0.2 g/L; 16.8% with Al_2O_3 at a concentration of 0.05 g/L; and 13% of SiO_2 at a concentration of 0.2 g/L [10]. However, under AC, the best improvement of AC-BDV does not exceed 7%, obtained with Fe_3O_4 at a concentration 0.4 g/L. The increase is 6% with Al_2O_3 at a concentration of 0.3 g/L. In some cases, the addition of NPs even reduces the AC-BDV of NE oil. A decrease of 15% is observed with SiO_2 at a concentration of 0.05 g/L [10]. Therefore, SE-based NFs are found to be more advantageous than NE-based NFs.

Based on the *p*-values, it is noticed that, with the exception of SE with Al_2O_3 at concentrations of 0.3 g/L and 0.4 g/L, and SE with SiO_2 at concentrations of 0.05 g/L and 0.2 g / L, the experimental findings with SE-based NFs comply with Weibull's probabilistic law. Similarly, with NE-based nanofluids with the exception of mixtures of NE/Fe_3O_4 (0.05 g/L), NE/Fe_3O_4 (0.4 g/L) and NE/SiO_2 (0.4 g/L).

Regarding the normal law, the hypothesis test and the *p*-values showed that the experimental results of SE-based NFs conform to the normal distribution with the exception of SE/Al_2O_3 (0.4 g/L), SE/SiO_2 (0.2 g/L) and SE/SiO_2 (0.3 g/L) NF samples. Similarly with NE-based NFs, all experimental data complies with the normal distribution except pure NE.

Regarding the processes involved in the presence of NPs, neither the polarity effect nor the development of streamers can be used to explain the experimental results. The experiments were carried out in a quasi-uniform electric field, even if the applied voltage waveform was a negative lightning impulse, due to symmetrical configuration of the electrodes (sphere-to-sphere electrode configuration). However, several mechanisms can be advanced to explain the effect of NPs, as discussed elsewhere [9].

However, the improvement in the dielectric withstand of esters in the presence of NPs may be due to (1) the fact that the NPs play the role of electron trap sites in the vicinity of the electrodes by reducing the number of electrons and consequently by raising the initiation threshold voltage of the streamer/discharge resulting in an increase of breakdown voltage; and (2) by reducing the number of electrons moving towards the opposite electrode (in the volume of the liquid), due to accumulation of electrons at the nanoparticle-surrounding liquid interfaces, forming a double layer, and thereby slowing down the development of the discharge leading to breakdown.

In the event that there is an excess of electrons on the surface of the nanoparticles and the double layer is saturated, the excess electrons which arrive later cannot be trapped; they will participate in the development of streamers, thus reducing the breakdown voltage. This may therefore explain the concept of optimal concentration; it is the threshold beyond which the electrons are no longer stopped. Thus, this process explains the concept of electron capture and release.

The improvement of the dielectric strength of esters-based nanofluids can be explained on the base of the influence of NPs on the streamer development as proposed by some authors such as Hwang et al. [27], Du et al. [28] and J. Miao et al. [29]. According to these authors, the NPs in particular the conducting ones, catch electrons in their movement under the effect of the electric field and accumulate them on their surfaces creating a local electric field which will oppose to the external electric field. The nanoparticles will act as "electron traps" inside the nanofluid. It results in a slowdown of the streamer propagation leading to the increase the breakdown voltage. Such a theory has been advanced for magnetic nanoparticles in the presence of lightning impulse voltage [27].

By using thermally stimulated current method (TSC) and pulse electroacoustic technique (PEA) for measuring the charge trap and transportation characteristics in pure transformer oil and transformer oil-TiO_2 based nanofluids submitted to positive impulse voltage, Du et al. [27] observed that electron shallow and trap density and charge decay rate are greatly increased in nanofluids. They concluded that fast electrons may be converted to slow electrons by electron trapping and de-trapping in shallow traps of nanofluids, resulting in an improved breakdown voltage compared to that of pure oil.

V. CONCLUSION

In this paper, the effects of magnetic (Fe_3O_4) and insulating (Al_2O_3 and SiO_2) NPs on the $U_{50\%}$ negative LI-BDV of natural and synthetic liquids (MIDEL 1204 and MIDEL 7131, respectively) were investigated. It is shown that the NPs improve the LI-BDV of both ester liquids, and that for each type of NP there is an optimal concentration of NP for which the $U_{50\%}$ LI-BDV is the highest. The LI-BDVs of investigated pure esters were found to be approximately twice those of the AC-BDVs.

The performed statistical analysis also showed that the experimental findings generally comply with normal and Weibull probabilistic laws.

The results presented in this work can have several applications such as the prediction of failures of nanofluids-filled apparatus in the event of lightning overvoltages and therefore the protection of high voltage equipment; or even in the techniques using repetitive pulse voltages such as electro-poration and treatment of some fluids, for example.

ACKNOWLEDGMENT

The authors extend their appreciation to RSSU at King Saud University for their technical support.

REFERENCES

[1] C. P. McShane, ''Natural and synthetic ester dielectric fluids: Their relative environmental, fire safety, and electrical performance,'' in *Proc. IEEE Ind. Commercial Power Syst. Tech. Conf.*, May 1999, pp. 1–8.

[2] D. Cherry, *Insulating Fluids: An Overview of Dielectric Fluids Used in Transformers*. Zürich, Switzerland: ABB Report, 2009.

[3] H. B. H. Sitorus, R. Setiabudy, S. Bismo, and A. Beroual, ''Jatropha curcas methyl ester oil obtaining as vegetable insulating oil,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 23, no. 4, pp. 2021–2028, Aug. 2016.

[4] T. V. Oommen, C. C. Claiborne, and J. T. Mullen, ''Biodegradable electrical insulation fluids,'' in *Proc. Electr. Insul. Conf. Electr. Manuf. Coil Winding Conf.*, Sep. 1997, pp. 465–468.

[5] H. Borsi and E. Gockenbach, ''Properties of ester liquid midel 7131 as an alternative liquid to mineral oil for transformers,'' in *Proc. IEEE Int. Conf. Dielectr. Liquids ICDL*, Jun. 2005, pp. 377–380.

[6] Y. Xuan and Q. Li, ''Heat transfer enhancement of nanofluids,'' *Int. J Heat Fluid Flow*, vol. 21, no. 1, pp. 58–64, 2000.

[7] S. U. S. Choi, ''Enhancing thermal conductivity of fluids with nanoparticles, developments and applications of non-Newtonian flows,'' Amer. Soc. Mech. Eng., New York, NY, USA, Tech. Rep. FED-vol. 231/MD-Vol. 66, 1995, pp. 99–105.

[8] L. Godson, B. Raja, D. M. Lal, and S. Wongwises, ''Enhancement of heat transfer using nanofluids—An overview,'' *Renew. Sustain. Energy Rev.*, vol. 14, pp. 629–641, Feb. 2010.

[9] U. Khaled and A. Beroual, ''AC dielectric strength of mineral oil-based Fe_3O_4 and Al_2O_3 nanofluids,'' *Energies*, vol. 11, no. 12, p. 3505, Dec. 2018.

[10] U. Khaled and A. Beroual, ''Statistical investigation of AC dielectric strength of natural ester oil-based Fe_3O_4, Al_2O_3, and SiO_2 nano-fluids,'' *IEEE Access*, vol. 7, pp. 60594–60601, 2019.

[11] U. Khaled and A. Beroual, ''AC dielectric strength of synthetic ester oil-based Fe_3O_4, Al_2O_3 and SiO_2 nanofluids—Conformity with Normal and Weibull distributions,'' *IEEE Trans. Dielectr. Electr. Insul.*, vol. 26, no. 6, pp. 625–633, 2019.

[12] *Green Power Transformers—GE Grid Solutions*. Accessed: 2016. [Online]. Available: http://www.GEGridSolutions.com

[13] V. Segal, A. Hjortsberg, A. Rabinovich, D. Nattrass, and K. Raj, ''AC (60 Hz) and impulse breakdown strength of a colloidal fluid based on transformer oil and magnetite nanoparticles,'' in *Proc. Conf. Rec. IEEE Int. Symp. Electr. Insul.*, Jun. 1998, pp. 619–622.

[14] Q. Wang, M. Rafiq, Y. Lv, C. Li, and K. Yi, ''Preparation of three types of transformer oil-based nanofluids and comparative study on the effect of nanoparticle concentrations on insulating property of transformer oil,'' *J. Nanotechnol.*, vol. 2016, no. 3, pp. 1–6, 2016.

[15] M. R., S. S. Kumar, and M. W. Iruthyarajan, ''A comparative investigation on effects of nanoparticles on characteristics of natural esters-based nanofluids,'' *Colloids Surf., Physicochem. Eng. Aspects*, vol. 556, pp. 30–36, Nov. 2018.

[16] Y. Lv, Y. Zhou, C. Li, Q. Wang, and B. Qi, ''Recent progress in nanofluids based on transformer oil: Preparation and electrical insulation properties,'' *IEEE Elect. Insul. Mag.*, vol. 30, no. 5, pp. 23–32, Sep. 2014.

[17] A. Elansezhiyan and S. Chandrasekar, ''Understanding the lightning discharge withstand and breakdown characteristics of nano SiO_2 modified mineral oil for transformer applications,'' *J. Adv. Chem.*, vol. 19, pp. 5200–5208, Dec. 2016.

[18] M. R. and S. Balaraman, ''Investigation on effects of different types of nanoparticles on critical parameters of nano-liquid insulation systems,'' *J. Mol. Liquids*, vol. 230, pp. 437–444, Mar. 2017.

[19] M. Makmud, H. Illias, C. Chee, and M. Sarjadi, ''Influence of conductive and semi-conductive nanoparticles on the dielectric response of natural ester-based nanofluid insulation,'' *Energies*, vol. 11, no. 2, p. 333, Feb. 2018.

[20] 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 Elect. Insul. Mag.*, vol. 28, no. 5, pp. 43–50, Sep. 2012.

[21] J. Chen, P. Sun, W. Sima, Q. Shao, L. Ye, and C. Li, ''A promising Nano-insulating-oil for industrial application: Electrical properties and modification mechanism,'' *Nanomaterials*, vol. 9, no. 5, p. 788, May 2019.

[22] W. Weibull, ''A statistical distribution function of wide applicability,'' *J. Appl. Mech.*, vol. 18, pp. 293–297, Sep. 1951.

[23] S. S. Shapiro and M. B. Wilk, ''An analysis of variance test for normality (complete samples),'' *Biometrika*, vol. 52, nos. 3–4, pp. 591–611, Dec. 1965.

[24] T. W. Anderson and D. A. Darling, ''Asymptotic theory of certain 'goodness of fit' criteria based on stochastic processes,'' *Ann. Math. Statist.*, vol. 23, no. 2, pp. 193–212, Jun. 1952.

[25] V.-H. Dang, A. Beroual, and C. Perrier, ''Comparative study of statistical breakdown in mineral, synthetic and natural ester oils under AC voltage,'' in *Proc. IEEE Int. Conf. Dielectric Liquids*, Jun. 2011, pp. 26–30.

[26] *The R Project for Statistical Computing*. Accessed: Jul. 2, 2018. [Online]. Available: www.r-project.org,version3.5.1

[27] J. G. Hwang, M. Zahn, F. M. O'Sullivan, L. A. A. Pettersson, O. Hjortstam, and R. Liu, ''Effects of nanoparticle charging on streamer development in transformer oil-based nanofluids,'' *J. Appl. Phys.*, vol. 107, no. 1, Jan. 2010, Art. no. 014310.

[28] Y. Du, Y. Lv, C. Li, M. Chen, J. Zhou, X. Li, Y. Zhou, and Y. Tu, ''Effect of electron shallow trap on breakdown performance of transformer oil-based nanofluids,'' *J. Appl. Phys.*, vol. 110, no. 10, Nov. 2011, Art. no. 104104.

[29] J. Miao, M. Dong, M. Ren, X. Wu, L. Shen, and H. Wang, ''Effect of nanoparticle polarization on relative permittivity of transformer oil-based nanofluids,'' *J. Appl. Phys.*, vol. 113, no. 20, May 2013, Art. no. 204103.

ABDERRAHMANE BEROUAL (Fellow, IEEE) is currently a Distinguished Professor at the École Centrale de Lyon, University of Lyon, France. He is also the Head of the High Voltage Group, AMPERE Laboratory – CNRS and a Scientific Expert at the Super Grid Institute. He was also responsible for the Master Research Program in electrical engineering from 2013 to 2015. He is the author/coauthor of more than 470 technical articles, including more than 210 refereed journal articles, five patents, two books, and six book chapters. He has supervised more than 45 Ph.D. theses. His main research interests include high voltage insulation, outdoor insulation, dielectric materials, long air gaps discharge and lightning, modeling of discharges, and composite materials. He is a Distinguished Visiting Professor of the U.K. Royal Academy of Engineering at Cardiff University and at King Saud University, Saudi Arabia. He was a recipient of the 2016 IEEE T. Dakin Award. From 1994 to 1998, he chaired the International Study Group on Streamer Propagation in Liquids of the IEEE – DEIS. He is a member of many advisory committees of international conferences, Technical Committee of the IEEE CEIDP, UF10 Technical Commission – MT30 of IEC, and an Associate Editor of the IEEE Transactions on Dielectrics and Electrical Insulation.

USAMA KHALED was graduated and received the M.Sc. degree in electrical engineering from Aswan University, Egypt, in 1998 and 2003, respectively, and the Ph.D. degree in electrical engineering from Cairo University, Egypt, and Kyushu University, Japan, through a joint scholarship, in 2010. Since 2000, he has been with the Department of Electrical Power Engineering, Faculty of Energy Engineering, Aswan University, working as a Teaching Assistant, an Assistant Lecturer, an Assistant Professor, and an Associate Professor. Since 2014, he had been an Assistant Professor at the College of Engineering, King Saud University, where he has been an Associate Professor, since 2018. His research interests include nanodielectric materials, applied electrostatics, and high voltage technologies.