

Received April 10, 2019, accepted May 4, 2019, date of publication May 8, 2019, date of current version May 21, 2019.

Digital Object Identifier 10.1109/ACCESS.2019.2915517

Statistical Investigation of AC Dielectric Strength of Natural Ester Oil-Based Fe_3O_4 , Al_2O_3 , and SiO_2 Nano-Fluids

USAMA KHALED^{1,2} AND ABDERRAHMANE BEROUAL³, (Fellow, IEEE)

¹Department of Electrical Engineering, College of Engineering, King Saud University, Riyadh 11421, Saudi Arabia

²Department of Electrical Engineering, Faculty of Energy Engineering, Aswan University, Aswan 81528, Egypt

³University of Lyon, Ecole Centrale de Lyon, CNRS, AMPERE Laboratory, UMR, CNRS 5005, 69134 Lyon, France

Corresponding author: Usama Khaled (ukhaled@ksu.edu.sa)

This work was supported by the King Saud University through the International Scientific Partnership Program (ISPP) under Grant ISPP-47.

ABSTRACT This paper deals with the effects of conductive and insulating nanoparticles at various sizes and concentrations on the ac dielectric strength of natural ester oil, namely, MIDEL 1204. The investigated nanoparticles are conductive (Fe_3O_4) and two insulating (Al_2O_3 and SiO_2). The measurements of breakdown voltages are achieved according to IEC 60156 standard. Experimental findings show that the investigated nanoparticles have not a significant influence on insulation performances of natural ester oil as it is the case with mineral oils and synthetic esters. The best improvement does not exceed 7%; it is obtained with Fe_3O_4 (50nm) at a concentration of 0.4 g/L and Al_2O_3 (13 nm) at a concentration of 0.05 g/L. In some cases, the addition of nanoparticles even reduces the dielectric strength of natural ester oil. Indeed, a decrease of 15% is observed with SiO_2 at a concentration of 0.05 g/L. The statistical analysis of experimental results is performed using two probabilistic functions, namely, normal and Weibull laws. It is shown that the breakdown voltage values of the investigated nano-liquids generally obey the normal and Weibull distributions. In addition, the breakdown voltage with a risk of 1% and 50% probability is deduced.

INDEX TERMS Breakdown, nano-fluids, natural ester, normal distribution, statistical analysis of experimental data, transformer oil, Weibull distribution.

I. INTRODUCTION

In an effort to safeguard the environment, both national and international policies demand/require that efforts be made in all social, economic and industrial sectors. Especially in the energy sector, the use of eco-friendly products for high voltage power transmission line and substation applications is very encouraged. So, for more than thirty years, many investigators are interested in the development of new materials with low impact on the environment as substitutes for those used till today in high voltage (HV) apparatus. Among HV applications, we are interested in, that of oil-filled apparatus such as power transformers. The most used insulating liquids in this component are mineral oils. Many researches have been conducted since three last decades on vegetable oils for the replacement of mineral oils. Vegetable oils indeed have better environmental performance (biodegradability and non-toxic), and higher fire safety guarantee than mineral and synthetic

ester oils [1]–[5]. Due to their high viscosity and oxidation instability (ageing instability), the use of vegetable oils is limited to non-breathing systems (for e.g., medium-voltage transformers). To circumvent these problems, vegetable oils are treated (esterification) so that their use can be extended. So, one can find today on the market some natural ester oils such as Biotemp, FR-3 and MIDEL 7131 [6], [7] that are currently used in many HV applications.

Among the qualities that must have an insulating oil (liquid) to be used in power transformers, a good dielectric strength and a good thermal transfer. One of the solutions to improve the heat transfer efficiency (cooling) could be the addition of nanoparticles. Indeed, nanofluids (NFs) possess better thermal conductivity, thermal diffusivity and convective heat transfer coefficient than the base fluids [8]–[12]. This cooling performance is one of the searched properties for transformer applications. So, according to some results reported in literature, the addition of conducting nanoparticles such as Fe_3O_4 greatly improve not only the thermal conductivity of mineral transformer oil but also its dielectric

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

strength [12], [13]–[15]. Similarly with semi-conducting and insulating nanoparticles even if the improvement of breakdown voltage is less important than that with Fe₃O₄ [16].

Contrary to mineral oils and synthetic ester, the influence of nanoparticles on the dielectric strength of natural ester (NE) does not seem to be so beneficial. The effect can even be reversed. Indeed, Peppas *et al.* [17] reported that the optimum AC breakdown voltage (BDV) of natural ester oil EnvirotempTM FR3 based oleic acid coated Fe₃O₄ nanofluids is obtained with 0.008% concentration what represents an improvement of about 20% with respect to the base oil (natural ester oil). Beyond this concentration, AC-BDV drastically drops in values well below the breakdown voltages of the base oil. According to the same authors [18], the AC breakdown voltage of NFs with SiO₂ nanoparticles is lower than that of natural ester. By adding Fe₃O₄ (conductive) and TiO₂ (semi-conducting) nanoparticles to another natural ester (NE) namely a highly-refined, bleached, deodorized palm oil (RBDPO), Makmud *et al.* [19] reported that the AC breakdown voltage of nanofluids is higher than that of pure NE. However, NE based – Fe₃O₄ nanofluid shows BDV higher than NE only at a lower concentration of these conductive nanoparticles. The AC-BDV of this NF decreases significantly when increasing Fe₃O₄ nanoparticles concentration. J. Li *et al.* [20] observed that positive and negative lightning impulse breakdown voltages of vegetable oil based – Fe₃O₄ nano-liquids increase by 37% and 12%; respectively.

From an economic point of view, the use of insulating liquids that are nanofluids with improved dielectric strength and heat transfer would enable to reduce the size, weight and costs of components (transformers), and to pass (transit) much higher power densities.

The purpose of this paper is to investigate the effects of conductive (Fe₃O₄) and insulating (Al₂O₃ and SiO₂) nanoparticles of various sizes and at different concentrations on AC breakdown voltage of natural ester oil namely MIDELE 1204. In first step are presented some characteristics of used nanoparticles, the procedure of preparation of nanofluid samples and the experimental arrangement for measurements of AC breakdown voltage; and then, the experimental results. An analysis of the conformity of the experimental results with Weibull and normal distributions is also performed. The physicochemical processes involved in breakdown phenomena of the investigated nanofluids are discussed.

II. EXPERIMENT

A. NANOFLUID SAMPLES PREPARATION

The investigated basic natural ester oil is MIDELE 1204. Table 1 gives its main characteristic parameters. Nanofluid samples are prepared using three types of nanoparticles: (1) conductive nanoparticles (Fe₃O₄) of 50 nm size; (2) insulating nanoparticles of SiO₂ with 10 to 20 nm size; and (3) insulating nanoparticles of Al₂O₃ with 13 and 50 nm sizes. Nanoparticles were supplied by Sigma-Aldrich; their purity is about 99.9%.

TABLE 1. Physicochemical properties of natural ester oil MIDELE 1204.

Property	MIDELE 1204
Density at 20°C (kg/dm ³)	0.92
Kinematic viscosity at 40°C (cSt)	37 cSt
Pour point (°C)	-31 °C
Flash point (°C)	>315 °C
Fire point (°C)	>350 °C
Total acid number (mg KOH/g)	>0.04
Water content (ppm)	100
Dissipation factor at 90°C	0.9 %

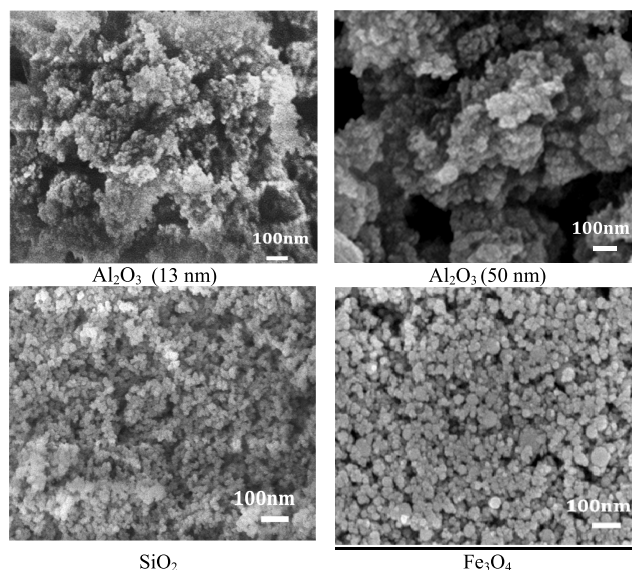


FIGURE 1. SEM (scanning electron microscope) images of various nanoparticles.

To check the distribution of particles (homogeneity), their shape and their composition at atomic percentage, similar methods and techniques as in previous works, were used [21], [22]. This includes particle size analyzer (NanoPlus, Particulate Systems - USA), scanning electron microscope (SEM) images and energy dispersive X-ray spectroscopy (EDS) analysis. From SEM images, one observes that these nanoparticle powders are more or less spherical and regular as shown in Figure 1; and the EDS analysis evidences the presence of each sample composition at an atomic percentage (Figure 2). Similarly, the natural ester-nanofluid samples were prepared in the same way as described elsewhere [21], [22].

B. BREAKDOWN MEASUREMENT

Fresh natural ester oil and natural ester oil-based nanofluid samples are tested according to the IEC 60156 standard [23] using an oil breakdown tester (Foster Oil Test 90 type) with a test cell of 500 ml volume containing two brass spherical electrodes of 12.5 mm diameter with an electrode gap of 2.50 ± 0.05 mm. The voltage is continuously applied at the electrodes at a uniform rise rate of 2 ± 0.2 kV/s until breakdown occurs. All measurements are performed under continuous stirring.

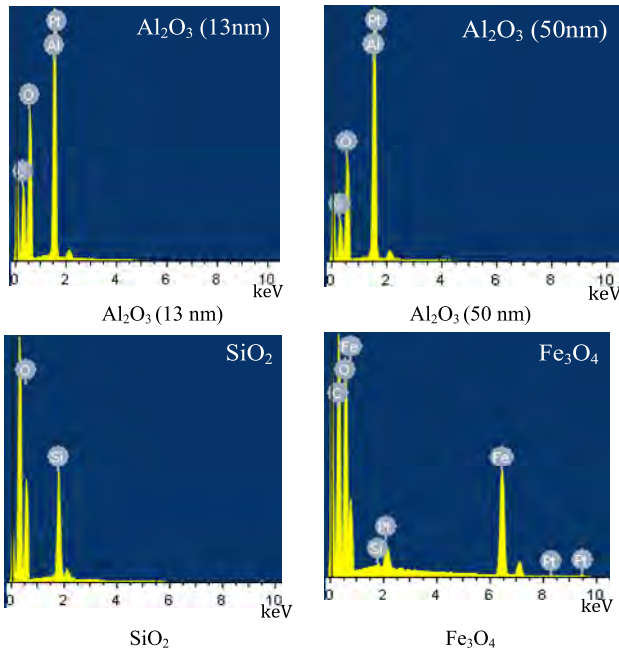


FIGURE 2. EDS images of various nanoparticles.

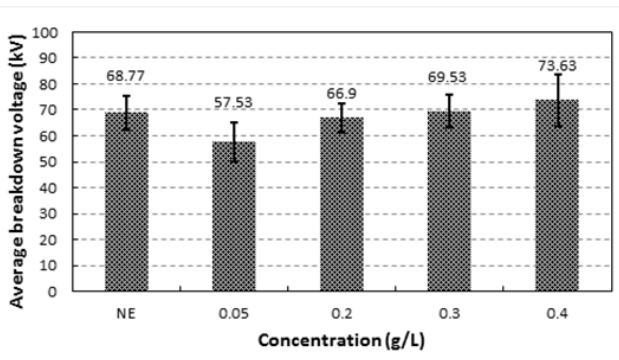


FIGURE 3. Average breakdown voltage of NE/Fe₃O₄ nano-fluids for different concentrations.

For the statistical investigations, two types of measurements were performed: (1) 5 series of six successive measurements each for the normal distribution analysis; let be 30 measurements in total; and (2) series of 16 (2⁴) measurements for the Weibull distribution, a reasonable number for this type of statistical analysis. The time delay between the successive measurements was 2 min. (according to IEC 60156 standard). This time is utilized as a stirring time to accomplish self-healing of nanofluid samples.

III. EXPERIMENTAL RESULTS

A. AVERAGE BREAKDOWN VOLTAGE

The average breakdown voltages (UBDV) of the different nanofluid samples, on the 5 series of six successive measurements each, are depicted in Figures 3 to 6. Table 2 summarizes the results for different concentrations of nanoparticles with their sizes. It comes out that, except at a concentration of Fe₃O₄, the breakdown voltage (BDV) of NF - Fe₃O₄ (50 nm)

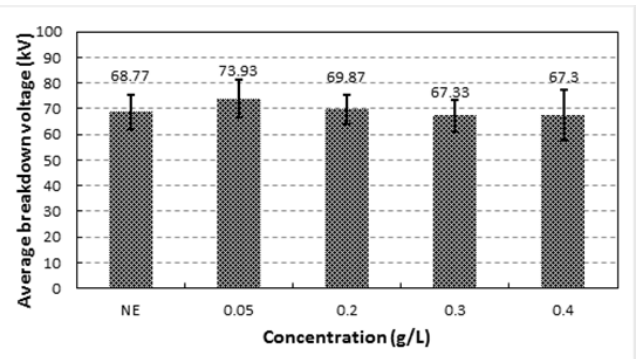


FIGURE 4. Average breakdown voltage of NE/Al₂O₃ (13nm) nano-liquids for different concentrations.

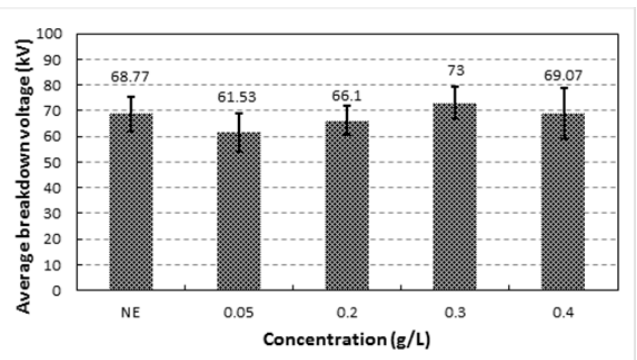


FIGURE 5. Average breakdown voltage of NE/Al₂O₃ (50nm) nano-liquids for different concentrations.

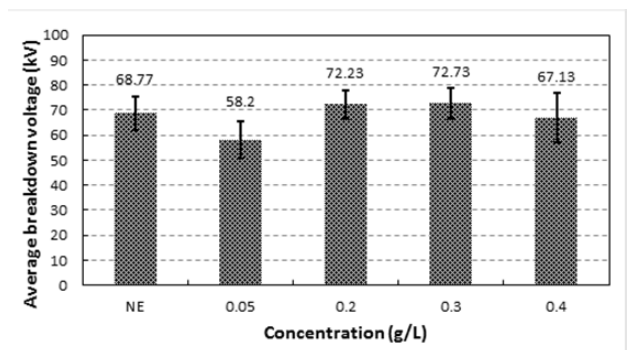


FIGURE 6. Average breakdown voltage of NE/SiO₂ nano-liquids for different concentrations.

is reduced regarding to the base natural ester oil; at 0.4 g/L, the breakdown voltage is enhanced by 7%.

With Al₂O₃ nanoparticles of 50 nm size at concentrations of 0.05 and 0.2 g/L, the breakdown voltages of NFs are lower than the base oil while they are slightly improved at 0.3 g/L (by 6%); with 0.4%, the improvement is insignificant (1%). For the same kind of nanoparticles with 13 nm size, there is a slight improvement at a concentration of 0.05% that is of 7%. With 0.2g/L, the enhancement is insignificant (1%); and the breakdown voltage even reduces at 0.3 and 0.4 g/L.

Concerning SiO₂ nanoparticles, the breakdown voltage is lower than that of the base oil at 0.05 g/L concentration.

TABLE 2. AC mean breakdown voltages of different nanofluids.

	Natural ester oil	Fe ₃ O ₄ (50nm)	Al ₂ O ₃ (50nm)	Al ₂ O ₃ (13nm)	SiO ₂ (50nm)
NE / 0.05 (g/L) NF					
BDV (kV)	68.77	57.53	61.53	73.93	58.20
Increment (%)	-	-16.34	-10.53	7.56	-15.37
NE / 0.2 (g/L) NF					
BDV (kV)	68.77	66.90	66.10	69.87	72.23
Increment (%)	-	-2.72	-3.88	1.59	5.03
NE / 0.3 (g/L) NF					
BDV (kV)	68.77	69.53	73.00	67.33	72.73
Increment (%)	-	1.11	6.15	-2.09	5.76
NE/ 0.4 (g/L) NF					
BDV (kV)	68.77	73.63	69.07	67.30	67.13
Increment (%)	-	7.07	0.44	-2.14	-2.38

Then, it increases a little bit by 5% at 0.2 g/L and 4% at 0.3 g/L; it somewhat decreases at 0.4 g/L.

B. STATISTICAL ANALYSIS OF EXPERIMENTAL DATA

In the following, an analysis of the statistical distribution of experimental results using the normal and Weibull distribution laws [24], [26] is achieved; these probabilistic distributions being the most used ones for analyzing the breakdown voltages of dielectrics. They constitute helpful tools in the design and maintenance of power apparatus by improving the predictability of the performance of the insulant. The normal distribution is a probability function that describes how the values of a variable are distributed; it is a symmetric distribution where most of the observations cluster around the central peak and the probabilities for values further away from the mean taper off equally in both directions. And the Weibull distribution enables to determine the probability of breakdown occurring at different percentages as well as the lifetime of insulant/equipment.

To check which distribution law, the experimental data would obey, two hypothesis tests were applied, namely Shapiro–Wilk test [26] and Anderson–Darling [27] test. For that purpose, one first calculates the probability *p* that measures the evidence against the null hypothesis using software R [28]. Then, the *p*-value is compared to a significant level α in order to decide if the data follow or not a normal distribution. Usually, one considers that a value of 5% for α is a reasonable value. In case of *p*-values equal or lower than α , the null hypothesis is rejected and hence the sample data do not belong to statistical distribution.

1) WEIBULL PROBABILITY OF DIFFERENT NANO-FLUID BASED ON NATURAL ESTER OIL

Figures 7 to 10 depict the Weibull plots of the AC breakdown voltage of natural ester-based Fe₃O₄, Al₂O₃ and SiO₂ nanofluids. In the inserts of these figures are given, the *p*-values, the Anderson–Darling (AD), the scale parameter related to the scattering of the data and indicating the degree of failure and the shape parameter; this later is equal to the

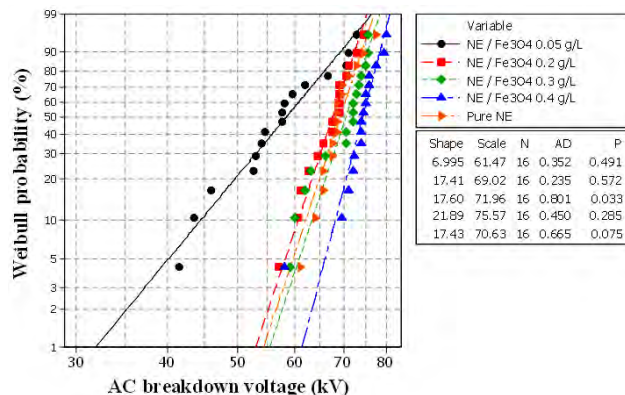


FIGURE 7. Weibull plot of the average reading of breakdown voltage of NE/Fe₃O₄ nanofluids.

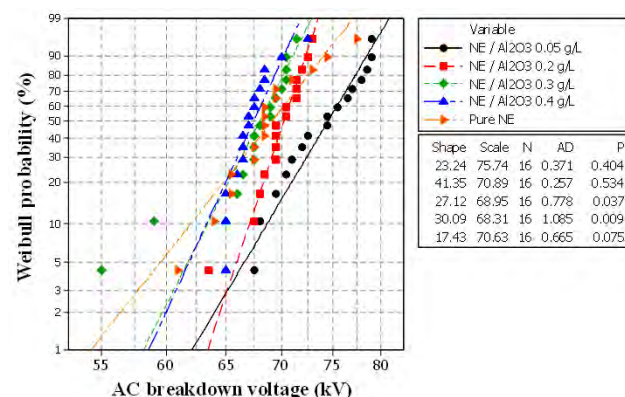


FIGURE 8. Weibull plot of the average reading of breakdown voltage of NE/Al₂O₃ (13nm) nanofluids.

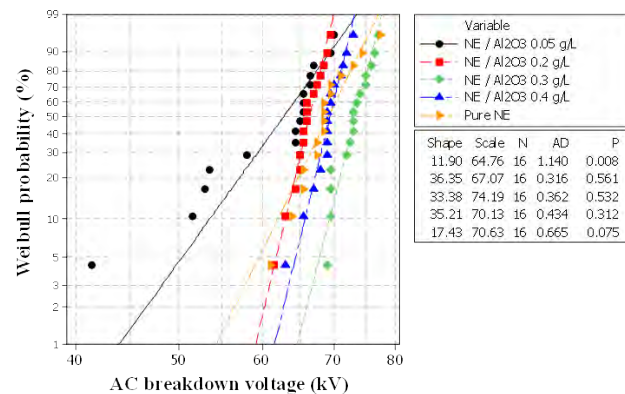


FIGURE 9. Weibull plot of the average reading of breakdown voltage of NE/Al₂O₃ (50nm) nanofluids.

slope of the line. The *p*-values and conformity of experimental results for Weibull distribution of investigated nanofluids are summarized in Table 3. It is noted that the experimental data generally conforms Weibull distribution except for some nanofluids (namely NE/Fe₃O₄ (0.3 g/L); NE/Al₂O₃ (13nm - 0.3 g/L), NE/Al₂O₃ (13nm - 0.4 g/L), NE/Al₂O₃ (50nm - 0.05 g/L)) for which the *p*-values are lower than 5%.

On the other hand, among the interesting and useful parameter from practical point of view, the AC breakdown voltage at

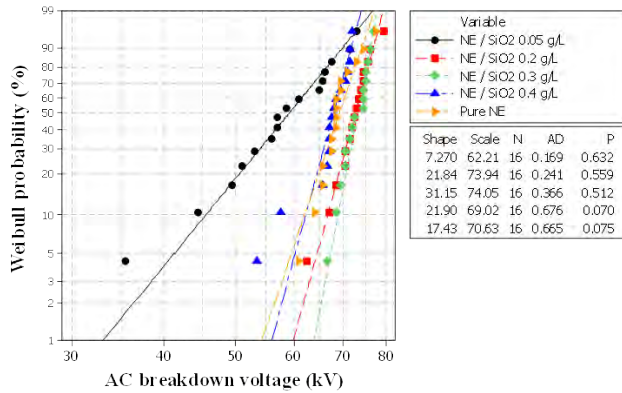


FIGURE 10. Weibull plot of the average reading of breakdown voltage of NE/SiO₂.

TABLE 3. Hypothesis test of conformity to weibull distribution of the average reading of breakdown voltage of nanofluids.

Sample	p-value	Conformity to Weibull distribution
Pure NE	0.075	Accepted
NE/Fe ₃ O ₄ (0.05 g/L)	0.491	Accepted
NE/Fe ₃ O ₄ (0.2 g/L)	0.572	Accepted
NE/Fe ₃ O ₄ (0.3 g/L)	0.033	Not Accepted
NE/Fe ₃ O ₄ (0.4 g/L)	0.285	Accepted
NE/Al ₂ O ₃ (13nm) (0.05 g/L)	0.404	Accepted
NE/Al ₂ O ₃ (13nm) (0.2 g/L)	0.534	Accepted
NE/Al ₂ O ₃ (13nm) (0.3 g/L)	0.037	Not Accepted
NE/Al ₂ O ₃ (13nm) (0.4 g/L)	0.009	Not Accepted
NE/Al ₂ O ₃ (50nm) (0.05 g/L)	0.008	Not Accepted
NE/Al ₂ O ₃ (50nm) (0.2 g/L)	0.561	Accepted
NE/Al ₂ O ₃ (50nm) (0.3 g/L)	0.532	Accepted
NE/Al ₂ O ₃ (50nm) (0.4 g/L)	0.312	Accepted
NE/ SiO ₂ (0.05 g/L)	0.632	Accepted
NE/ SiO ₂ (0.2 g/L)	0.559	Accepted
NE/ SiO ₂ (0.3 g/L)	0.512	Accepted
NE/ SiO ₂ (0.4 g/L)	0.070	Accepted

1% and 50% cumulative probabilities. Indeed, the breakdown voltage at 1% cumulative probability that is the lowest possible breakdown voltage constitutes useful information about the reliability of insulating liquid; it is helpful for the design of insulating system; and the breakdown voltage at 50% cumulative probability is the mean value. Table 4 summarizes the AC breakdown voltage at 1% and 50% breakdown probabilities for investigating nanofluids.

Contrary to mineral oil and synthetic ester - based nanofluids where the improvement of AC breakdown voltages was very interesting when adding Fe₃O₄, magnetic/conducting nanoparticles, the enhancement of natural ester-based nanofluids is less important. Indeed, for the same kinds of nanoparticles (Fe₃O₄, Al₂O₃ and SiO₂), the best improvement of mineral oil is obtained with Fe₃O₄, nanoparticles at a concentration of 0.4 g/L; it can exceed twice that of pure mineral oil [21]. For synthetic ester, namely MIDEAL 7131, the best improvement is of 50%; it is also obtained with the same type of nanoparticles (i.e.; Fe₃O₄) at

TABLE 4. AC breakdown voltage at different breakdown probabilities of 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				
Fe₃O₄ Nanoparticles									
1	54.3	31.8	-41.4	53.0	-2.4	55.4	2.0	61.3	12.9
50	69.2	58.3	-15.8	67.6	-2.3	70.5	1.9	74.3	7.4
Al₂O₃ (13 nm) Nanoparticles									
1	54.3	62.1	14.4	63.4	16.8	58.2	7.2	58.6	7.9
50	69.2	74.6	7.8	70.3	1.6	68.0	-1.7	67.5	-2.5
Al₂O₃ (50 nm) Nanoparticles									
1	54.3	44.0	-19.0	59.1	8.8	64.6	19.0	61.5	13.3
50	69.2	62.8	-9.2	66.4	-4.0	73.4	6.1	69.4	0.3
SiO₂ Nanoparticles									
1	54.3	32.8	-39.6	60.0	10.5	63.8	17.5	55.9	2.9
50	69.2	59.0	-14.7	72.7	5.1	73.2	5.8	67.9	-1.9

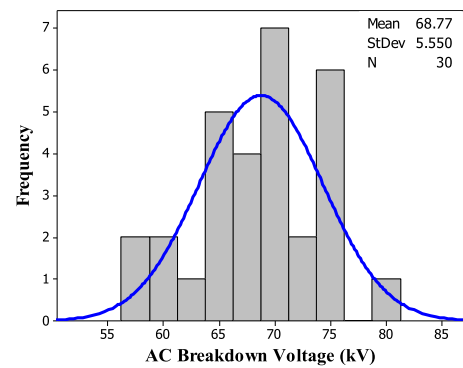


FIGURE 11. Histogram of pure NE.

a concentration of 0.4 g/L [22]. While with natural ester - based nanofluids; the best result is obtained with Fe₃O₄ (50nm) at a concentration 0.4 g/L and for Al₂O₃ (13nm) at a concentration of 0.05 g/L for which the improvement does not exceed 7%. At low concentration of Fe₃O₄ nanoparticles (0.05 and 0.2 g/L), the breakdown voltage of nanofluids decreases. It is even observed a decrease by 15% with SiO₂ at a concentration of 0.05 g/L.

2) HISTOGRAM AND NORMAL DISTRIBUTION OF AC BREAKDOWN VOLTAGE OF INVESTIGATED NANO-FLUIDS

The histograms of the distribution of breakdown voltage of the tested samples are shown in Figures 11 to 15.

Such a representation allows us to detect eventual anomalies in the distribution of breakdown voltage in different ranges of voltages. Table 5 summarizes the p- values for the experimental data of investigated nanofluids and the conformity or not to normal distribution. It is observed that the experimental data obey the normal distribution except for some nanofluids (namely NE/Fe₃O₄ (0.3 g/L),

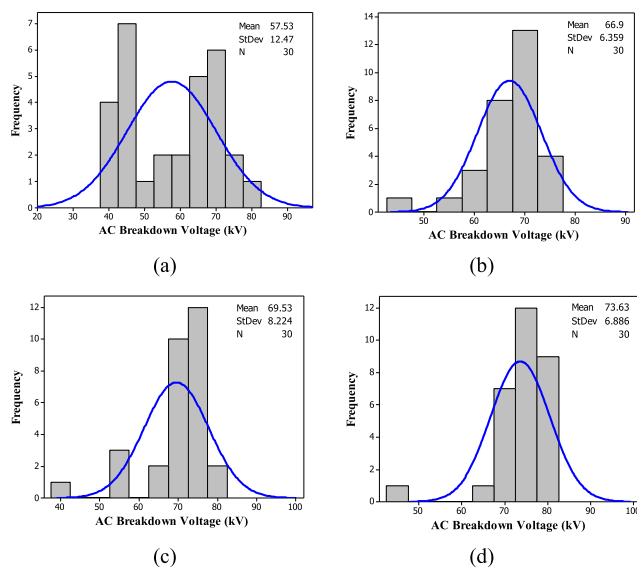


FIGURE 12. Histogram of NE/Fe₃O₄ of different concentrations. (a) 0.05 g/L. (b) 0.2 g/L. (c) 0.3 g/L. (d) 0.4 g/L.

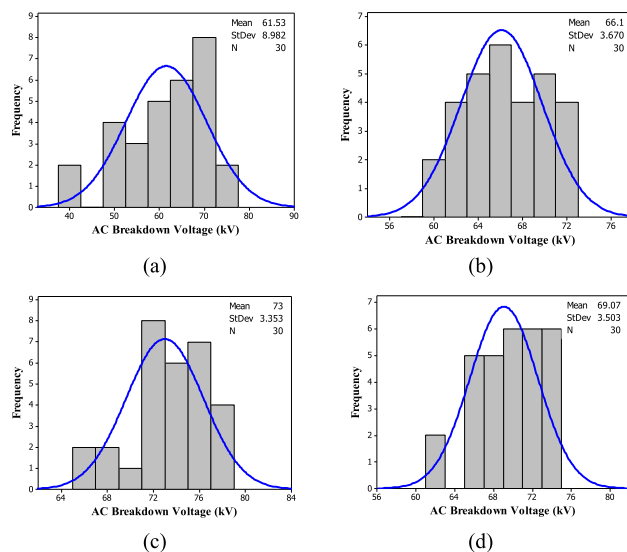


FIGURE 14. Histogram of NE/Al₂O₃ (50nm) of different concentrations. (a) 0.05 g/L. (b) 0.2 g/L. (c) 0.3 g/L. (d) 0.4 g/L.

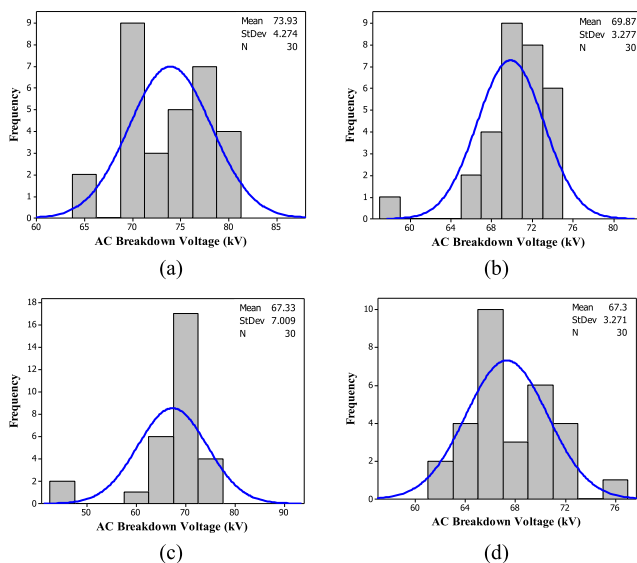


FIGURE 13. Histogram of NE/Al₂O₃ (13nm) of different concentrations. (a) 0.05 g/L. (b) 0.2 g/L. (c) 0.3 g/L. (d) 0.4 g/L.

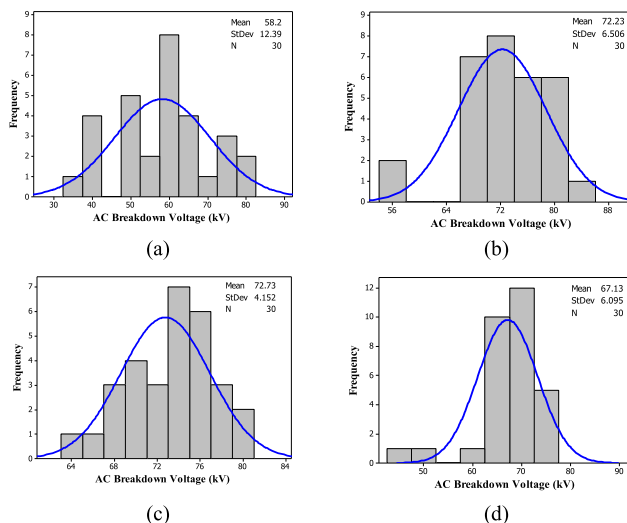


FIGURE 15. Histogram of NE/SiO₂ of different concentrations. (a) 0.05 g/L. (b) 0.2 g/L. (c) 0.3 g/L. (d) 0.4 g/L.

NE/Fe₃O₄ (0.4 g/L), NE/Al₂O₃ (13nm - 0.3 g/L), NE/Al₂O₃ (50nm - 0.05 g/L) and NE/SiO₂ (0.4 g/L)) for which the *p*-values are lower than 0.05. To evaluate whether or not there is problem with non-normality, one can also use the measure of skewness and kurtosis similarly as conducted in previous work [22] using statistical packages such as SPSS (*Statistical Package for the Social Sciences*) [29].

Note that the AC breakdown voltage at 1% and 50% cumulative probabilities deduced from the histograms of nanofluid samples are close to those deduced from the Weibull plots as shown in Table 6.

IV. DISCUSSION

It appears from the experimental results that from the point of view of dielectric strength, natural ester oil based-Fe₃O₄,

Al₂O₃ and SiO₂ nano-fluids are not so interesting as is the case with mineral oil and synthetic ester with the same nanoparticles. Indeed, the improvement is too weak compared with the results obtained with mineral oil or synthetic ester [21], [22]. It does not exceed 7%, with the investigated nanoparticles. The breakdown voltage can even decrease as it is the case with SiO₂ nanoparticles for which one observes a reduction of 15% at a concentration of 0.05 g/L.

Such an increase with natural ester – based nanofluid for a given concentration of nanoparticles and then a decrease for another concentration has been reported by Peppas *et al.* [17], [18]. These authors observed that the addition of 0.008% wt./wt. of commercial Fe₃O₄ powder or oleate-coated colloidal Fe₃O₄ at a concentration of 0.012% wt./wt. increases the breakdown voltage of natural ester oil Envirotemp™ FR3 by 20%. However, with further addition

TABLE 5. Hypothesis test of conformity to normal distribution of the average reading of breakdown voltage of various nanofluids.

Sample	p-value	Conformity to Normal distribution
Pure NE	0.371	Accepted
NE/Fe ₃ O ₄ (0.05 g/L)	0.643	Accepted
NE/Fe ₃ O ₄ (0.2 g/L)	0.430	Accepted
NE/Fe ₃ O ₄ (0.3 g/L)	0.010	Not Accepted
NE/Fe ₃ O ₄ (0.4 g/L)	0.010	Not Accepted
NE/Al ₂ O ₃ (13nm) (0.05 g/L)	0.465	Accepted
NE/Al ₂ O ₃ (13nm) (0.2 g/L)	0.181	Accepted
NE/Al ₂ O ₃ (13nm) (0.3 g/L)	0.005	Not Accepted
NE/Al ₂ O ₃ (13nm) (0.4 g/L)	0.159	Accepted
NE/Al ₂ O ₃ (50nm) (0.05 g/L)	0.005	Not Accepted
NE/Al ₂ O ₃ (50nm) (0.2 g/L)	0.728	Accepted
NE/Al ₂ O ₃ (50nm) (0.3 g/L)	0.290	Accepted
NE/Al ₂ O ₃ (50nm) (0.4 g/L)	0.125	Accepted
NE/ SiO ₂ (0.05 g/L)	0.742	Accepted
NE/ SiO ₂ (0.2 g/L)	0.381	Accepted
NE/ SiO ₂ (0.3 g/L)	0.252	Accepted
NE/ SiO ₂ (0.4 g/L)	0.005	Not Accepted

TABLE 6. AC breakdown voltage at different breakdown probabilities of nanofluids (from normal distribution).

Breakdown voltage probability (%)	0.05 g/L				0.2 g/L				0.3 g/L				0.4 g/L			
	Breakdown voltage (kV)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Breakdown voltage (kV)	Increment (%)	Breakdown voltage (kV)	Breakdown voltage (kV)	Increment (%)	
	Fe₃O₄ Nanoparticles															
1	54.3	28.5	-47.5	52.1	-4.1	50.4	-7.2	57.6	6.1	69.2	17.6	73.6	4.4	73.6	6.4	
50	69.2	57.5	-16.9	66.9	-3.3	69.5	0.4	73.6	6.4	73.6	0.0	73.6	0.0	73.6	0.0	
	Al₂O₃ (13 nm) Nanoparticles															
1	54.3	64.0	17.9	62.2	14.5	51.0	-6.1	59.7	9.9	69.2	27.0	67.3	-2.7	67.3	-2.7	
50	69.2	73.9	6.8	69.9	1.0	67.3	-2.7	67.3	-2.7	69.2	0.0	67.3	-2.7	67.3	-2.7	
	Al₂O₃ (50 nm) Nanoparticles															
1	54.3	40.6	-25.2	57.6	6.1	65.2	20.1	60.9	12.2	69.2	14.3	67.3	-2.7	67.3	-2.7	
50	69.2	61.5	-11.1	66.1	-4.5	73.0	5.5	69.1	-0.1	69.2	0.0	67.3	-2.7	67.3	-2.7	
	SiO₂ Nanoparticles															
1	54.3	29.4	-45.9	57.1	5.2	63.1	16.2	53.0	-2.4	69.2	15.1	67.3	-2.7	67.3	-2.7	
50	69.2	58.2	-15.9	72.2	4.3	72.7	5.1	67.1	-3.0	69.2	0.0	67.3	-2.7	67.3	-2.7	

of nano-Fe₃O₄, the breakdown voltage significantly reduces. They attributed this to the decreasing inter-particle distance of the nano-Fe₃O₄, which starts forming conductive paths above a threshold value.

While other investigators reported that the AC breakdown voltage of Palm Fatty Acid Ester (PFAE) oil-based nanofluids with a concentration of 0.01 g/L of Fe₃O₄ conductive nanoparticles gives a significant improvement of dielectric performance compared to pure PFAE, whereby the AC breakdown voltage increases by 43% [30]. Menlik *et al.* [31] also observed that the silica-coated titanium (ST TiO₂) particles at a concentration of 0.25% improves the average value of

breakdown voltage of ENVITRAFOL (a biodegradable oil) by about 33% (from 60 kV for pure liquid to 80.15 kV); the tests were performed according to IEC 60156.

The fact that, these nanoparticles do not always improve the dielectric strength of natural ester, has been also reported by others [32]. By considering virgin linseed and virgin castor, Chetty *et al.* [32] observed that the addition of 5 wt% of silica aerogel powder nanoparticles increased the streamer activity.

Therefore, and unlike mineral oils and synthetic esters for which the additions of some specific nanoparticles (especially Fe₃O₄) improve significantly the breakdown strength, there is no unanimous conclusion about the effect of nanoparticles on natural esters. It is therefore difficult to propose a mechanism that could explain the variation (increase/decrease) of the dielectric strength of natural esters - based nanofluids, as has been the case for mineral oils and synthetic esters - based nanofluids [21], [22] in which case a mechanism of trapping electrons by nanoparticles or of slowing the development/propagation of streamers were evoked.

Therefore, it would also be difficult to envisage the use of this type of nanofluids in high voltage applications (power transformers), at least based on MIDEL 1204. This would bring no added value compared to vegetable oil alone.

V. CONCLUSION

In this work, the insulation performances of natural ester oil – MIDEL 1204, in the presence of three kinds of nanoparticles (namely conductive (Fe₃O₄) and two insulating (Al₂O₃ and SiO₂) nanoparticles), were investigated. It was shown that the effect of nanoparticles on the breakdown voltage of this natural ester is not evident /complex. Indeed, the investigated nanoparticles have a less effect on the breakdown voltage of natural ester than that observed with mineral oils and synthetic esters. The best improvement does not exceed 7%; it is obtained with Fe₃O₄ (50nm) at a concentration 0.4 g/L and Al₂O₃ (13nm) at a concentration of 0.05 g/L. In some cases, the addition of nanoparticles reduces the dielectric strength of natural ester oil; a decrease of 15% was observed with SiO₂ at a concentration of 0.05 g/L. Thus, the use of this kind of nanofluids in high voltage applications, at least based on MIDEL 1204, could arise some problems.

It is also shown that the values of breakdown voltage values of the investigated nano-liquids generally obey the normal and Weibull distributions.

REFERENCES

- [1] T. V. Oommen, C. C. Claiborne, and J. T. Mullen, "Biodegradable electrical insulation fluids," in *Proc. Elect. Insul. Elect. Manuf. Coil Winding Conf.*, Rosemont, IL, USA, Sep. 1997, pp. 465–468.
- [2] 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.*, Sparks, NV, USA, May 1999 p. 8.
- [3] T. V. Oommen, "Vegetable oils for liquid-filled transformers," *IEEE Elect. Insul. Mag.*, vol. 18, no. 1, pp. 6–11, Jan. 2002.
- [4] S. Lindren, "Environmentally acceptable transformer fluids," EPRI, Palo Alto, CA, USA, Tech. Prog. 1000438, 2000.
- [5] D. Cherry, "Insulating fluids: An overview of dielectric fluids used in transformers," ABB, Zürich, Switzerland, Nov. 2009.

- [6] 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. Dielectric Liquids*, Coimbra, Portugal, Jun./Jul. 2005, pp. 377–380.
- [7] *MIDEL Selection Guide*, M&I Mater. Ltd., Manchester, U.K., Mar. 2017.
- [8] Y. Xuan and Q. Li, "Heat transfer enhancement of nanofluids," *Int. J. Heat Fluid Flow*, vol. 21, no. 1, pp. 58–64, 2000.
- [9] J. A. Eastman and S. U. S. Choi, "Anomalous increased effective thermal conductivities of ethylene glycol-based nanofluids containing copper nanoparticles," *Appl. Phys. Lett.*, vol. 78, no. 6, p. 718, 2001.
- [10] S. Choi and J. Eastman, "Enhancing thermal conductivity of fluids with nanoparticles," in *Developments and Applications of Non-Newtonian Flows (FED)*, vol. 231/MD-vol. 66, D. Siginer and H. Wang, Eds. New York, NY, USA: ASME, 1995, pp. 99–105.
- [11] S. U. S. Choi and Z. G. Zhang, "Anomalous thermal conductivity enhancement in nanotube suspensions," *Appl. Phys. Lett.*, vol. 79, p. 2252, Aug. 2001.
- [12] 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.
- [13] R. Mdavan and B. Sujatha, "Investigation on effects of different types of nanoparticles on critical parameters of nano-liquid insulation systems," *J. Mol. Liquids*, vol. 230, pp. 437–444, 2017.
- [14] V. Segal, A. Hjortsberg, A. Rabinovich, D. Natrass, and K. Raj, "AC (60 Hz) and impulse breakdown strength of a colloidal fluid based on transformer oil and magnetite nanoparticles," in *Proc. Int. Symp. Elect. Insul.*, Arlington, VA, USA, Jun. 1998, pp. 619–622.
- [15] Y. Lv *et al.*, "Nanoparticle effect on dielectric breakdown strength of transformer oil-based nanofluids," in *Proc. Annu. Rep. Int. Conf. Elect. Insul. Dielectric Phenomena*, Shenzhen, China, Oct. 2013, pp. 680–682.
- [16] Y. Z. Lv, Y. Zhou, C. R. 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./Oct. 2014.
- [17] G. D. Peppas, V. P. Charalampakos, E. C. Pyrgioti, M. G. Danikas, A. Bakandritsos, and I. F. Gonos, "Statistical investigation of AC breakdown voltage of nanofluids compared with mineral and natural ester oil," *IET Sci. Meas. Technol.*, vol. 10, no. 6, pp. 644–652, Sep. 2016.
- [18] G. D. Peppas, V. P. Charalampakos, E. C. Pyrgioti, A. Bakandritsos, A. D. Polykrati, and I. F. Gonos, "A study on the breakdown characteristics of natural ester based nanofluids with magnetic iron oxide and SiO₂ nanoparticles," in *Proc. Int. Conf. High Voltage Eng. Appl. (ICHVE)*, Athens, Greece, Sep. 2018, pp. 10–13.
- [19] M. Z. H. Makmud, H. A. Illias, C. Y. Chee, and M. S. 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, 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./Oct. 2012.
- [21] U. Khaled and A. Beroual, "AC dielectric strength of mineral oil-based Fe₃O₄ and Al₂O₃ nanofluids," *Energies*, vol. 11, no. 12, p. 3505, 2018.
- [22] U. Khaled and A. Beroual, "AC dielectric strength of synthetic ester based Fe₃O₄, Al₂O₃ and SiO₂ nanofluids—Conformity with normal and Weibull distributions," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 26, no. 2, pp. 625–633, Apr. 2019.
- [23] *Insulating Liquids—Determination of the Breakdown Voltage at Power Frequency—Test Method*, document IEC 60156:1995, 1995.
- [24] W. Weibull, "A statistical distribution function of wide applicability," *J. Appl. Mech.*, vol. 18, no. 3, pp. 293–297, 1951.
- [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*, Trondheim, Norway, Jun. 2011, pp. 1–4.
- [26] 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, 1965.
- [27] T. W. Anderson and D. A. Darling, "Asymptotic theory of certain 'goodness of fit' criteria based on stochastic processes," *Ann. Math. Stat.*, vol. 23, no. 2, pp. 193–212, 1952.
- [28] (2018). *The R Project for Statistical Computing*. [Online]. Available: www.r-project.org
- [29] D. George and P. Mallery, *SPSS for Windows Step by Step: A Simple Guide and Reference, 11.0 Update*, 10th ed. Boston, MA, USA: Pearson, 2010.
- [30] M. S. Mohamad, H. Zainuddin, S. A. Ghani, and I. S. Chairul, "AC breakdown voltage and viscosity of palm fatty acid ester (PFAE) oil-based nanofluids," *J. Elect. Eng. Technol.*, vol. 12, no. 6, pp. 2333–2341, 2017.
- [31] V. Mentlik, P. Trnka, J. Hornak, and P. Totzauer, "Development of a biodegradable electro-insulating liquid and its subsequent modification by nanoparticles," *Energies*, vol. 11, no. 3, p. 508, 2018. doi: [10.3390/en11030508](https://doi.org/10.3390/en11030508).
- [32] L. Chetty, I. W. Serukenya, and N. M. Ijumba, "Vegetable oil based liquid nanocomposite dielectric," *South Afr. J. Sci.*, vol. 109, nos. 1–2, pp. 1–6, 2013.



USAMA KHALED received the bachelor's degree and the M.Sc. degree in electrical engineering from Aswan University, Egypt, in 1998 and 2003, respectively, the Ph.D. degree in electrical engineering from Cairo University, Egypt, and the Ph.D. degree in electrical engineering from 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,

where he has been a Teaching Assistant, an Assistant Lecturer, an Assistant Professor, and an Associate Professor. His research interests include nanocomposite materials, applied electrostatics, nanofluids, and high voltage technologies. He has been an Assistant Professor, since 2014, and an Associate Professor, since 2018, with the College of Engineering, King Saud University.



ABDERRAHMANE BEROUAL (F'11) is currently Distinguished Professor with the Ecole Centrale de Lyon, University of Lyon, France. He is currently the Head of the High Voltage Group, AMPERE Lab., CNRS, and a Scientific Expert with the SuperGrid Institute. He is also responsible for the master's research program in electrical engineering. His main research interests include high voltage insulation, outdoor insulation, dielectric materials, long air gaps discharge and lightning, and the modeling of discharges and composite materials. He has supervised over 45 Ph.D. theses. He has authored or coauthored over 450 technical papers including more than 190 refereed journal papers, 2 books, and 6 book chapters. He held five patents. He is a Distinguished Visiting Professor with the U.K. Royal Academy of Engineering, Cardiff University, and King Saud university, Saudi Arabia. He is a member of many Advisory Committees of International Conferences, Technical Committee of the IEEE CEIDP, and UF10 Technical Commission—MT30 of IEC. From 1994 to 1998, he was the Chair of the International Study Group on Streamer Propagation in Liquids of the IEEE—DEIS. He was a recipient of the 2016 IEEE T. Dakin Award. He is an Associate Editor of the IEEE TDEI.

• • •