

Article

Statistical Analysis of AC Dielectric Strength of Natural Ester-Based ZnO Nanofluids

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Abstract: Due to environmental concerns and increased energy demand, natural esters are among the alternatives to mineral oils in transformers. This study examines the electrical behavior of natural ester-based ZnO nanofluids at different concentrations in the range of 0.05–0.4 g/L. AC breakdown voltages are measured in a horizontally positioned sphere–sphere electrode system according to IEC 60156 specifications. The measurement data are analyzed using Weibull and normal distribution functions. Breakdown voltages with 1%, 10% and 50% probability are also estimated, these probabilities being of great interest for the design of power electrical components. Experimental results show that AC breakdown voltage increases with the concentration of ZnO nanoparticles, except for the concentration of 0.05 and 0.4 g/L of ZnO. Moreover, breakdown voltages at 1% and 10% probability increase by 22.7% and 13.2% when adding 0.1 g/L ZnO to natural ester, respectively.

Keywords: naturel ester oil; nanofluids; zinc oxide; AC breakdown voltage; Weibull distribution; normal distribution



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1. Introduction

Insulating liquids are widely used in insulating systems for high voltage (HV) electrical components such as transformers, cables, power capacitors, reactors, circuit breakers, bushings and tap changers [1]. Insulation and heat transfer are among the main requirements that these liquids have to ensure in HV power transformers, these latter being indispensable components for the transmission and distribution electrical energy systems. In HV transformers, 75% of total faults are caused by insulation problems. These transformer failures reduce the expected life of transformers by almost half [2]. Note that the insulation system consists of insulating oil (transformer oil) and solid insulating (paper and pressboard) [3]. The main functions of transformer oils are electrical insulation, protection of solid insulation against air and moisture, improvement of solid insulation performance by penetrating cellulose, protection against corrosion and cooling [3].

The most commonly used insulating liquids in transformers are mineral oils. These latter have been marketed and used since the end of the 19th century for their relatively low cost, dielectric and cooling properties, and compatibility with cellulose-based solid insulation materials and availability [4]. Despite these common advantages, mineral oils have significant disadvantages such as flammability, low biodegradability, low moisture tolerance and corrosive sulphur compounds [5]. Low flash and fire temperatures raise heat protection problems and therefore require measures such as fire safety, firewalls and deluge systems [6]. Mineral oils consisting of different hydrocarbon compounds are a by-product of the oil industry. Oil resources will eventually be depleted; some estimates predict that oil shortages may emerge in the middle of the twenty-first century [4]. This turns out to be an important threat to the insulating liquids industry, where several billion liters are used [1]. Another environmental problem of mineral oils is their low biodegradability,

below 30% for 28 days [7]. In the event of a leak or spill after an accident, it pollutes the soil and groundwater and turns into an important threat to humans and the ecosystem.

In order to avoid the problems related to these disadvantages, many studies of alternative insulating liquids were launched over more than forty years [8]. Among the requirements that these alternative liquids are expected to meet are a high dielectric strength, a good heat transfer, improved fire safety, good sustainability, environmentally friendly and extended service life [2,9]. The protection of the environment, which has become a demand/requirement nowadays, is a deciding criterion for alternative transformer oils [8]. Environmentally friendly transformer oil is defined by high biodegradability and low toxicity [2].

Alternatives to mineral oils include many types of insulating liquids in the categories of high molecular weight hydrocarbons, synthetic and natural esters [1,10]. Natural esters obtained from plants such as rapeseed, soybean, sunflower, olive, palm and jatropha, consist mainly of triglycerides, which contain unsaturated fatty acids [1,4,11]. The advantages of natural esters compared to mineral oils are high flash and fire temperatures, almost completely biodegradability, non-toxicity, high dielectric strength and high moisture tolerance [7,9]. Natural esters with fire temperatures above 300 °C can be used in applications where there is a risk of fire without taking special safety precautions [1]. Power system equipment using these less flammable oils can be positioned at lower separation/safety distances [6]. Natural esters that dissolve in nature within 28 days with a rate of over 95% also successfully meet environmental requirements [4]. These oils have higher dielectric strength than mineral oils. Due to this feature, binary mixtures with mineral oils increase the breakdown voltage in power system equipment such as transformers [7]. These properties have made it so that natural esters have been used in transformer and capacitor applications since the early 1990s [11]. The use of natural esters is increasing especially in coastal areas where transformer oil can be contaminated with water and in applications where fire risk can cause great economic damage [1,11].

The major disadvantages of natural esters are increased dielectric dissipation factor (loss factor, $\tan \delta$) at high temperatures, high pour temperature, high viscosity and poor oxidation stability [12,13]. The pour temperature is the lowest temperature at which liquid materials can maintain their fluidity properties. High pour temperature turns into a disadvantage in terms of the use of natural esters in cold climatic conditions. These disadvantages can lead to exceeding standard limits in terms of thermal, loss and electrical aspects. For example, Fofana et al. [14] have found that when the ratio of natural ester in the binary mixture with mineral oils is more than 50%, the density and viscosity parameters of the mixture exceed the standard limits. Hermetically sealed applications that prevent contact of natural esters with moisture and oxygen are widely recommended to overcome these problems [13]. In addition, reducing the ratio of unsaturated fatty acids with esterification processes of these oils is another alternative approach [11].

To circumvent and resolve the drawbacks that natural esters present, and in general to improve the thermal and electrical characteristics of insulating liquids, nanoparticles (NP) were introduced into these liquids in the mid-1990s [15]. Originally, nanoparticle-added fluids or nanofluids (NFs) aimed mainly at improving thermal characteristics such as diffusivity, conductivity, convective coefficient and heat transfer [16]. In addition to the fact that they improve thermal properties, some nanoparticles also make it possible to increase the dielectric strength of fluids. Therefore, NFs constitute ideal alternative insulators for oil-filled high voltage applications [17]. The advantages of NP-added transformer oils include better AC, DC and impulse breakdown performances, better partial discharge characteristics, less sensitivity to moisture, prolonged insulation and transformer life, increased thermal conductivity and better cooling of transformers [2].

Different types of NPs are used in the preparation of these nanofluids. Nanoparticles can be classified into three main categories: Conductive, semi-conductive and non-conductive. These classifications, which are defined in terms of electrical behavior, make it easier to define and discuss the breakdown mechanisms [16]. The most commonly

used NPs in transformer oils are titanium dioxide (TiO_2), iron oxide (Fe_3O_4), aluminium oxide (Al_2O_3), silicon dioxide (SiO_2) and zinc oxide (ZnO), respectively [18]. Apart from these, NPs such as copper oxide (CuO), fullerene (C_{60}) and aluminium nitride (AlN) are also studied [1,19]. The AC and positive impulse breakdown voltages of these NP-added transformer oils could be improved by a factor of up to 50% [2,19,20]. This rate of increase in breakdown voltages differs depending on the type, size, shape and concentration of the NPs and the type of transformer oil [17]. The addition of Fe_3O_4 to mineral oil can more than double the AC breakdown voltage [16].

Hanai et al. [21] observed that the AC breakdown voltages of ZnO-based mineral oils increased by up to 8.3% compared to pure mineral oil. Bakruthen et al. [22] found that this increase is 40.6% at 0.075% concentration for a different mineral oil. Chen et al. [20] observed that AC, positive and negative polarity impulse breakdown voltages increased up to 30.2%, 18.9% and 35.8%, respectively, in FR3 oil with 0.4 g/L ZnO.

The breakdown voltage characteristic and mechanism of ZnO, a semiconductor nanoparticle such as TiO_2 , in insulating liquids has not been widely studied. Considering that almost 30% of the studies on natural ester-based nanofluids in the literature use TiO_2 [18], the examination of semiconductor ZnO nanoparticle doped nanofluids offers a potential innovation.

This study examines the AC breakdown voltage characteristics of natural ester-based ZnO nanofluids at concentrations of 0.05 to 0.4 g/L; the measurements are conducted according to the IEC 60,156 standard. The experimental results are analyzed with Weibull and normal distribution functions, and probabilities of 1%, 10% and 50% breakdown stresses are deduced.

2. Experiment

2.1. Preparation of Nanofluids

The natural ester MIDELE eN 1204 transformer oil used in this study is based on rapeseed. The physicochemical properties of this oil are shown in Table 1. The ZnO nanoparticles used in the preparation of the nanofluid are supplied from PlasmaChem GmbH. The average diameter of these spherical particles is 25 ± 3.5 nm and the density is 5.606 g/cm^3 at 20°C with 99.5% purity.

Table 1. Physicochemical properties of MIDELE eN 1204.

Property	MIDELE eN 1204
Density at 20°C (g/cm^3)	0.92
Kinematic viscosity at 40°C (mm^2/s)	8
Pour temperature ($^\circ\text{C}$)	−31
Flash Point ($^\circ\text{C}$)	>315
Fire Point ($^\circ\text{C}$)	>350
Total acid number (mg KOH/g)	0.04
Water content (ppm)	50
Dissipation factor at 90°C	<0.1

Two different methods are used in the preparation of NFs. In the one-step method, nanoparticles are synthesized and dispersed simultaneously in the base fluid. This method does not include the drying, storage and transportation of nanofluids. Therefore, agglomeration is minimized and fluid stability is improved [10]. Due to the high cost of large-scale production with this method, the two-step method is preferred for nanofluids based on transformer oils [2]. In this method, the fluid is firstly mixed with nanoparticles using a magnetic stirrer as depicted in Figure 1. After this step, surfactant is added to this solution and nanofluid is produced by ultrasonication [23]. In order to avoid the agglomeration problem, measurements are taken after the preparation of the nanofluid. Nanofluids prepared by the two-step method exhibit a homogeneous characteristic for several months

without any agglomeration problems [10]. This method can be used on an industrial scale for almost all nanofluids. In this study, oleic acid is used as surfactant.

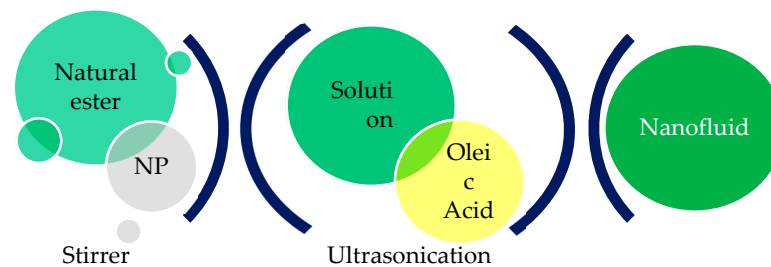


Figure 1. Diagram of two-step method for preparation of nanofluids (NFs).

Due to the high surface energies and attractive/repulsive forces of nanoparticles, the NF can become unstable. Surfactant reduces the surface tension of the fluid and increases the immersion of NPs [2]. This mechanism is also defined as steric stabilization. Oleic acid is the most widely used in NFs prepared with transformer oils. Apart from oleic acid, long-chain hydrocarbons such as hexadecyl trimethyl ammonium bromide (CTAB), sorbitan esters and sodium dodecyl sulphate (SDS) are also used, but rarely [10,23].

In this study, natural ester is purified by using a micro membrane filter and vacuum pump. ZnO NPs of five different concentrations ranging from 0.05 to 0.4 g/L are added to this fluid. Then, each sample is mixed with a magnetic stirrer for 30 min. Oleic acid is added to this solution and an ultrasonication process is applied for 2 h with an ultrasonic homogenizer. This process ensures that the NPs are homogeneously dispersed in the fluid and remain stable without aggregations/clusters. Sonics Vibra-cell sonicator used in ultrasonication has 500 W power rating, 20 kHz frequency and 60% amplitude. This ultrasonication process is applied in periods of 20 min with a 10 min waiting time between each to prevent the nanofluid from overheating. In order to eliminate possible humidity and micro air bubbles that develop during the preparation of the nanofluid, the nanofluid is kept in the oven and then under vacuum at a pressure of 1.0 Pa for 24 h.

2.2. AC Breakdown Measurement

AC breakdown voltages of pure Midel eN-1204 and these natural ester-based ZnO nanofluids are measured using the BAUR DTA 100C, according to the IEC60156 [24] using a 400 mL test cell, 12.5 mm diameter electrodes horizontally positioned at a 2.50 ± 0.05 mm electrode gap. The rest time of each sample is 30 min in order to eliminate gas bubbles in the test cell. The voltage is increased with a rise rate of 2 kV/s until breakdown occurs. The time delay between each breakdown is 2 min and the number of measured breakdown voltages in each set is 6. In order to have sufficient data for statistical analysis, five series of six measurements each, i.e., a total of 30 measurements, are carried out on each type of nanofluid sample [5,8]. After the measurement, the test cell and electrodes are cleaned with ethanol. After this stage, it is washed with hot water at a temperature of 60–80 °C and dried in an oven at 60 °C for one hour. This procedure is compatible with approaches in similar measurement studies reported in the literature [4,5].

The AC breakdown voltage characteristics are analyzed with Weibull and normal distribution functions, and the withstand voltage levels at 1%, 10% and 50% are determined.

3. Results and Discussions

The measurement results taken to check the conformity or not of their distribution with the Weibull law and the normal law are presented in Figure 2. The mean and standard deviation of these measurements are calculated using Equations (1) and (2), respectively:

$$\bar{U} = \frac{1}{n} \sum_{i=1}^n U_i \quad (1)$$

$$\sigma = \sqrt{\frac{1}{n} \sum_{i=1}^n (U_i - \bar{U})^2} \tag{2}$$

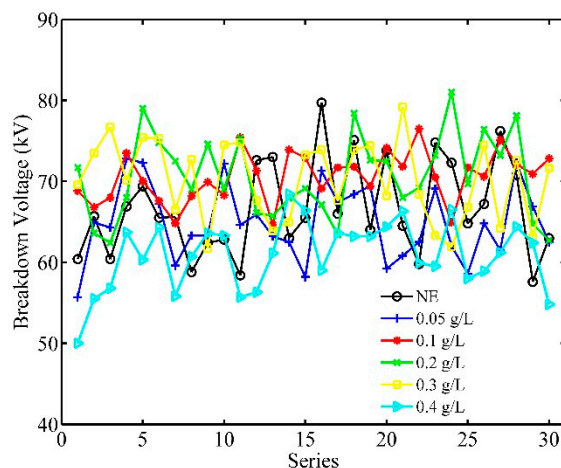


Figure 2. Distribution of AC breakdown voltages of natural ester (NE) and NFs.

It is noticed that the AC breakdown voltages of natural ester (NE) are reduced by 2.7% and 8.7% for nanofluids with a concentration of 0.05 and 0.4 g/L ZnO, respectively. This characteristic changes for 0.1, 0.2 and 0.3 g/L ZnO and increases by 5.8%, 5.8% and 5.1%, respectively.

Khaled and Beroual [8] examined the same natural ester-based Fe₃O₄, Al₂O₃ and SiO₂ NFs and observed that the best improvement in breakdown voltages did not exceed 7%. The breakdown voltage was reduced by about 15% in the 0.05 g/L-added SiO₂ nanofluid [8].

The mean and standard deviation range of these breakdown voltage measurements at different concentrations are given in Figure 3. The ratio of standard deviation to mean breakdown voltage measurements is 5.9% to the maximum for pure naturel ester and 3.1% to the minimum for 0.1 g/L ZnO concentration. The growth of this ratio is linearly related to the difference between the measurements in each concentration set.

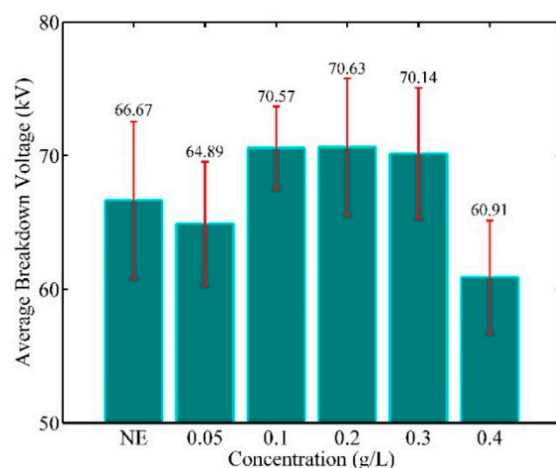


Figure 3. Average breakdown voltages of NE and ZnO nanofluids for different concentrations.

Histogram charts of NE and nanofluids are given in Figure 4. These charts include breakdown frequency at different voltage levels, mean value of breakdown voltage and standard deviation. In terms of average breakdown voltage, 0.1, 0.2 and 0.3 g/L ZnO NFs perform better than NE.

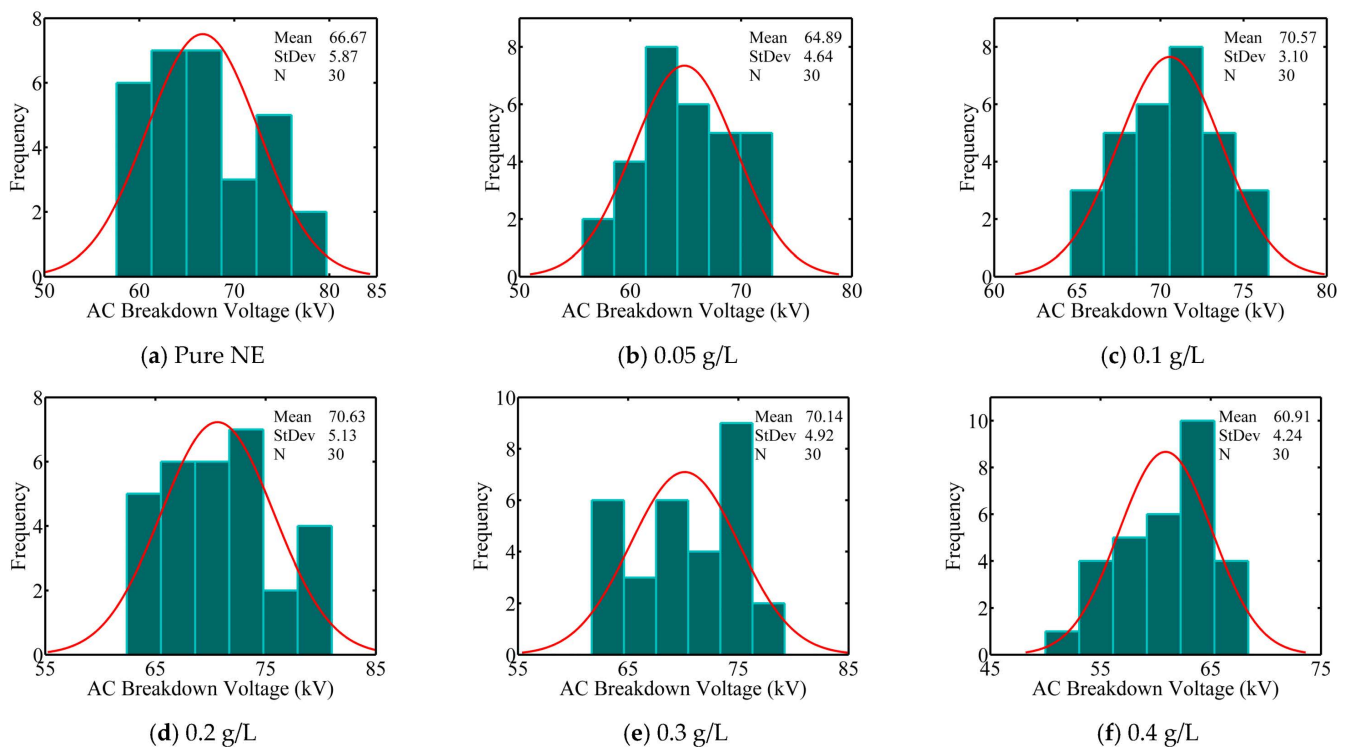


Figure 4. Histograms of NE and ZnO nanofluids for different concentrations.

In order to statistically analyze the probability of breakdown voltage by adhering to these measurements, the Weibull and normal distribution should be tested using a hypothesis. In the test of this hypothesis, which questions the distribution of measurements in the 5% significance level ($\alpha = 0.05$), Anderson–Darling and Shapiro–Wilk tests are used for Weibull and normal distributions, respectively [7,8].

The Anderson–Darling normality test can examine measurement data without grouping and is very sensitive to distributions in the tail region rather than the median [25]. The Shapiro–Wilk test is a regression-correlation based test using a sequential sample. This test, in which the normality of the samples is tested, is consistent in all alternative datasets up to 50 samples [26]. The p -value is the probability of making an error in testing the hypothesis that the measurement data conforms with the statistical law [7].

A hypothesis is accepted if the p -value obtained in these tests is greater than the significance level. Under the condition that the hypothesis is accepted, the distribution of the measurements is defined as a statistical distribution and different probability levels can be estimated [27].

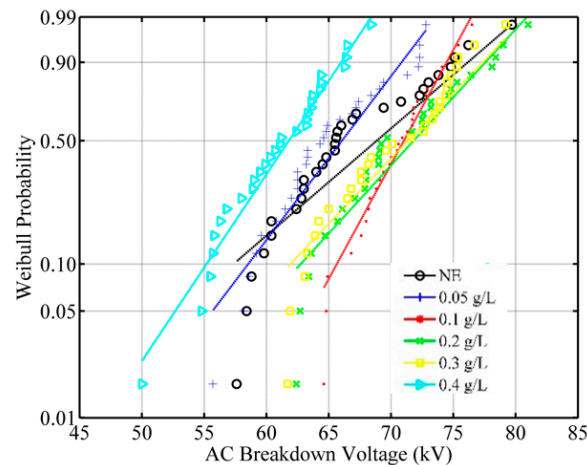
W , which is also defined as the test statistic, is evaluated differently for both tests. The hypothesis is rejected due to a too large W value in the Anderson–Darling test. In order for the hypothesis to be accepted, W should be below 1.5786 at the 0.05 significance level [25]. In the Shapiro–Wilk test, in order for the hypothesis to be accepted at the 95% confidence interval, W should be in the range of 0.9303–1.0000 depending on the number of samples used [26]. The test statistics provide the necessary conditions for both tests and the normal distribution hypothesis is accepted.

The Weibull distribution of the measurements given in Figure 2 is examined using the Anderson–Darling test. According to this test, the distributions of measurements for natural ester and natural ester-based ZnO nanofluid samples are within the acceptable significance level, see Table 2. The acceptance of hypothesis tests of conformity for all samples allows the analysis of the breakdown voltage characteristics using the Weibull distribution function.

Table 2. Hypothesis test of conformity to Weibull distribution of NE and NFs.

	W	p-Value	Conformity of Weibull Distribution
NE	0.5449	0.1611	Accepted
0.05 g/L	0.3345	0.5149	Accepted
0.1 g/L	0.1782	0.9109	Accepted
0.2 g/L	0.2860	0.6007	Accepted
0.3 g/L	0.7278	0.0516	Accepted
0.4 g/L	0.3842	0.3957	Accepted

The probability curves due to the Weibull distribution are shown in Figure 5. In the 50% probability region, 0.1 to 0.3 g/L ZnO nanofluids have similar AC breakdown voltage characteristic. In this region, the AC breakdown voltage of 0.05 and 0.4 g ZnO nanofluids worsens. In the 10% probability region, the best breakdown voltage is in the 0.1 g/L ZnO nanofluid sample.

**Figure 5.** Weibull probability of NE and ZnO nanofluids for different concentrations.

The same sets of measurements are used to examine the compliance with the normal distribution function. The statistical distributions of these measurements are examined with the Shapiro–Wilk test and the hypothesis that the normal distribution is distributed in the significance level ($\alpha = 0.05$) is accepted in all samples, see Table 3.

Table 3. Hypothesis test of conformity to normal distribution of NE and NFs.

	W	p-Value	Conformity of Normal Distribution
NE	0.9516	0.1866	Accepted
0.05 g/L	0.9626	0.3608	Accepted
0.1 g/L	0.9756	0.7005	Accepted
0.2 g/L	0.9654	0.4231	Accepted
0.3 g/L	0.9385	0.0830	Accepted
0.4 g/L	0.9670	0.4601	Accepted

Using these normal distribution functions in which experimental measurements are analyzed, 1%, 10% and 50% probability breakdown voltages can be estimated, see Table 4. The statistical nature of the breakdown voltages complicates the design of power system equipment. In order to overcome these difficulties, the withstand voltages calculated as a statistical parameter are defined with different possibilities. The withstand voltage of the insulation is not the average value of the breakdown voltage, but as a statistical variable, a low probability of breakdown voltage, such as 1% or 10% [28]. These critical risk levels for design safety are widely studied in the literature [7,28,29]. The 1% probability of

breakdown voltage is a safety factor in the design of electrical equipment and is defined as the voltage limit for operation in the safety margin [7,28].

Table 4. AC breakdown withstand voltages at different probabilities for NE and NFs.

BDV Probability	NE BDV (kV)	Type of NFs	BDV (kV)	Change (%)
%1	51.17	0.05 g/L	54.16	5.9
		0.1 g/L	62.78	22.7
		0.2 g/L	57.92	13.2
		0.3 g/L	56.83	11.1
		0.4 g/L	51.02	−0.3
%10	58.52	0.05 g/L	59.12	1.0
		0.1 g/L	66.27	13.2
		0.2 g/L	63.72	8.9
		0.3 g/L	62.94	7.5
		0.4 g/L	55.43	−5.3
%50	67.55	0.05 g/L	65.20	−3.5
		0.1 g/L	70.55	4.4
		0.2 g/L	70.85	4.9
		0.3 g/L	70.45	4.3
		0.4 g/L	60.85	−9.9

These breakdown probabilities are also considered as withstand stress levels. NFs other than that with 0.4 g/L ZnO outperform natural ester in terms of 1% withstand voltage probability. Specifically, a 22.7% increase in 0.1 g/L sample significantly improves the withstand voltage performance for this critical parameter. Similarly, this value increases for 0.2 and 0.3 g/L ZnO samples; the rate of increase is 13.2% and 11.1%, respectively. In the withstand voltage where there is a 10% probability of breakdown, the 0.1 g/L sample shows the best performance as in the previous probability level. The increase rate in this withstand voltage for 0.1 g/L nanofluid is 13%.

The electrical insulation characteristics of nanofluids with ZnO additives are improved compared to the natural ester in terms of withstand breakdown voltages at 1% and 10% probabilities.

Mechanisms of breakdown characteristics of nanofluids are not clearly defined and remain the subject of controversy. The electrical conductivity of nanoparticles seems to be an important parameter in the explanation of this mechanism. Conductive nanoparticles capture the rapidly moving electron in the fluid and turn into slow negatively charged nanoparticles. Streamer propagation slows down and therefore the breakdown voltage increases with this mechanism [16]. Conductive and nonconductive nanoparticles trap electrons by charge induction polarization, respectively. These scavenger nanoparticles reduce free electrons moving in the fluid [5].

ZnO is a semiconductor nanoparticle that traps high mobility electrons. It slows down the electrons responsible for streamer development with trapping and de-trapping processes [2]. In nanofluids using semiconductor nanoparticles, the surface trap density and charge dissipation velocity are 2.5 and 4 times higher compared to pure transformer oil, respectively [30]. With the effect of these mechanisms, AC, DC and impulse breakdown voltages can increase by 20% compared to pure oil [30]. The capture of electrons by scavenger nanoparticles increases the initial threshold voltage of the streamer, and therefore more energetic breakdown mechanisms emerge [16,27]. Due to the semiconductor property of the ZnO, the mechanism defined as bridging or tunnelling develops when the nanoparticle density exceeds a certain concentration [19,21]. In this case, layers adjacent to nanoparticles separate insulating oil and these layers act as a conductor in a very high electric field [20]. Approximately 10% reduction of breakdown voltage at a concentration of 0.4 g/L can be explained by this mechanism.

Breakdown voltages in nanofluids using ZnO nanoparticles can increase by 8.3% to 40.6% in different concentrations and fluids [21,22]. The positive and negative impulse breakdown voltage of natural ester-based ZnO nanofluids increased by 19.8% and 35.8%, respectively, for the 0.4 g/L ZnO concentration [20]. Despite the findings in these studies, the AC breakdown characteristics and withstand voltages of natural ester-based ZnO nanofluids are presented in detail with this study. The increase in AC breakdown and withstand voltages can be up to 5.8% and 22.7% for 0.1 g/L ZnO concentration, respectively.

The AC breakdown voltage averages of mineral oils commonly used in power transformers measured using the same method are 38.5 kV [16], 39.0 kV [28] and 51.6 kV [7]. The AC breakdown voltage measurement averages of synthetic esters under the same conditions are 47.0 kV [28] and 60.03 kV [5]. The electrical insulation characteristic of the natural ester, whose average AC breakdown voltage is measured as 66.67 kV, has a better performance than mineral oils and synthetic esters. This insulating performance can be improved with the addition of ZnO nanoparticles and provides a more reliable insulating medium alternative for the transformer.

The flash and fire points of natural esters are higher than mineral oils and synthetic esters. Due to these properties, the performance of power transformers using natural esters continues without deterioration even when exposed to high temperatures [6].

4. Conclusions

The main findings obtained in this study can be summarized as follows:

- In natural ester-based ZnO nanofluids, breakdown voltages decrease for 0.05 and 0.4 g/L concentrations and increase for 0.1 to 0.3 g/L concentrations. The best improvement in breakdown voltage is of about 5%; it is obtained with a concentration of 0.1 g/L ZnO.
- Breakdown voltage data of all samples comply with normal distribution. Using these distribution functions, risks of 1%, 10% and 50% probabilities of breakdown voltages are calculated. The best improvement in 1% probability withstand voltage is in 0.1 g/L ZnO nanofluid. At this concentration, the probability of breakdown voltage increases by 22.7%. The improvement at concentrations of 0.2 and 0.3 g/L for the same probability is 13.2% and 11.1%, respectively. For 10% probability, the best performance is also in the 0.1 g/L concentration. At this probability, the development of the breakdown voltage is 13%.
- This increase in breakdown voltages of ZnO-added natural esters provides the opportunity to design power system equipment, especially transformers, in smaller dimensions and to meet the increasing demand.
- It is thought that the critical value of the amount of nanoparticles is exceeded at a concentration of 0.4 g/L. Increasing nanoparticle concentration beyond this value reveals the implication of a tunnelling/bridging mechanism that leads to a reduction in breakdown voltages.

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