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Influence of Fullerene Nanoparticles on AC and LI Breakdown Voltages of Natural Ester

Hidir Duzkaya
Dept. of Electrical - Electronic Eng.
Gazi University
Ankara, Turkey
hduzkaya@gazi.edu.tr

Abderrahmane Beroual
Laboratoire Ampère UMR-CNRS 5005
Ecole Centrale de Lyon – University of Lyon
Ecully, France
Abderrahmane.Beroual@ec-lyon.fr

Abstract—This study aims to investigate the AC and lightning impulse (LI) breakdown voltage performance of natural ester-based nanofluids enriched with fullerene (C₆₀) nanoparticles, which stand out with their long-term stability and superior dielectric properties. The AC and LI breakdown voltages of these natural ester-based nanofluids with five different concentrations of fullerene (C₆₀) are measured 30 times and the compatibility of these measurements with the statistical distribution is checked with the Anderson-Darling test. The AC breakdown voltage performances of 0.3 g/L and 0.4 g/L C₆₀ nanofluids are 5.1% and 7.8% better, respectively, than natural ester. The LI breakdown voltage performance of 0.1 g/L C₆₀ nanofluid is 8.2% better than natural ester. After accepting the conformity of these measurements with the statistical distribution, AC and LI withstand voltages are calculated at 1%, 10% and 50% breakdown voltage probabilities. The withstand voltages of both AC and LI nanofluids are better than natural ester in almost all samples at 1% breakdown voltage probability. The results show that natural ester-based C₆₀ nanofluids can meet the electrical strength requirements of power system equipment with better AC and LI breakdown voltage performance.

Keywords—natural ester, fullerene, AC breakdown, lightning impulse breakdown, Weibull distribution.

I. INTRODUCTION

Insulating fluids are used in many power system equipment applications such as power transformers, tap changers, cables and capacitors [1]. These fluids, which are also defined as transformer oil due to their widespread use in power transformers, increase the electrical and thermal performance of these equipment. Some of the main functions of these fluids are protecting the solid insulation material against moisture and air, increasing the dielectric strength, protecting against corrosion and cooling [2].

Mineral oils have conventionally been used in the insulating fluid industry for over a hundred years [1]. These oils have key advantages such as relatively good dielectric and cooling performance, low cost, availability and compatibility with solid insulating material in transformers [3]. The main shortcomings of mineral oils are relatively low flash/fire points, low moisture tolerance, corrosive sulfide decomposition products and low biodegradability [4]. These thermal, electrical and environmental disadvantages make the search for alternatives a necessity for the billion-liter insulating fluid industry [1]. Alternative dielectric fluids need to meet requirements such as increase fire safety, improve heat transfer and dielectric strength, increase the service life of power system equipment, be environmentally friendly and sustainability [5].

Although there are synthetic esters and high molecular weight hydrocarbons among the alternatives of mineral oils, natural esters stand out in terms of critical parameters such as high dielectric strength, environmental impact and availability [2]. Natural esters are produced from vegetable seeds such as olive, canola, coconut, rapeseed, sunflower and palm [1]. The main advantages of these esters over mineral oils are almost twice the AC dielectric strength and flash/fire points, complete biodegradability and high moisture tolerance [3]. These advantages of natural esters have been exploited since the early 1990s as binary mixtures with mineral and synthetic oils for potential more compact designs and retro-filling applications of power transformers [6, 7].

The disadvantages of natural esters include high pour temperatures, oxidation instability, increased dissipation factor at high temperatures, and high viscosity compared to mineral oils [3, 8]. These disadvantages can cause the electrical and thermal limits determined by the standards to be exceeded in applications where natural esters are used [6, 8].

Nanoparticles have been used to improve the thermal and electrical characteristics of fluids since the 1990s. The thermal properties of these nanoparticle-added nanofluids such as conductivity, heat transfer, diffusivity and convective coefficient can be improved compared to the base fluid [9]. The dielectric strength of these nanofluids can also improve depending on the nanoparticle and the base fluid [10]. Nanoparticles such as Al₂O₃, Fe₃O₄, SiO₂, TiO₂ and ZnO are commonly used to improve the electrical and thermal properties of transformer oils. The AC and positive impulse breakdown voltages of these nanoparticles-added nanofluids can increase up to 50% depending on the type, concentration and shape of the nanoparticle and the type of fluid [9-11].

Research on fullerene-added nanofluids is a relatively new topic in the literature. Nadolny and Dombek [11] determined that the heat transfer factor of 0.1 g/L C₆₀-added synthetic ester-based nanofluid could be improved by 10% and other important parameters did not change compared to the base fluid. Chen *et al* [2019] reported that the AC, positive and negative LI breakdown voltages of mineral oil-based fullerene nanofluid increased by 18%, 7.5% and 8.3%, respectively, compared to base oil. The AC and LI breakdown voltages of natural ester-based fullerene nanofluids increase by up to 10% compared to pure natural ester in spherical electrode geometry [13].

This study aims to investigate the AC and LI breakdown stress measurements of natural ester-based fullerene nanofluids at five different concentrations. The compatibility of these measurements with the statistical distribution is checked with the Anderson-Darling test and the Weibull

distribution function is calculated for all samples. The withstand voltages of natural esters and nanofluids at 1%, 10% and 50% breakdown voltage probabilities are determined by using these distribution functions.

II. EXPERIMENTAL PROCEDURE

A. Preparation of Nanofluids

In this study, Midel eN1204 natural ester, which is a mixture of rapeseed and canola oils, is the base fluid. The fullerene nanoparticle has a purity of 98% and an average particle size of 21 nm. There are one- and two-step methods in the literature for the preparation of nanofluids. The one-step method is based on the principle of simultaneous synthesis and mixing of nanoparticles in a fluid [14]. Although more homogeneous nanofluids are produced with this method, it is not preferred in transformer oils due to its cost and incompatibility with large-scale industrial production. The two-step method, which is preferred in the preparation of transformer oil-based nanofluids, is based on the principle of producing and mixing the fluid and nanoparticle separately, see Fig. 1.

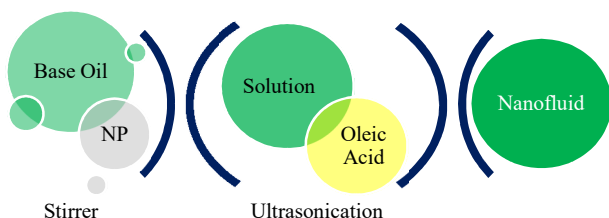


Fig. 1. Schematic diagram of two-step method for preparation of NFs [3]

Natural ester-based nanofluids with five different fullerene nanoparticle concentrations between 0.05 g/L and 0.4 g/L are prepared for this study. The detailed procedure followed during the preparation of these nanofluids can be found in the authors' previous work [13].

B. Measurement Procedure

AC breakdown voltages of natural ester and fullerene nanofluids are measured using the BAUR DTA 100C measuring system according to the IEC60156 standard [15]. LI breakdown voltages are measured using a two-stage Marx generator (200 kV-2 kJ) producing standard impulse voltage characteristics with 1.2/50 μ s. For each sample, the AC and LI breakdown voltages are measured 30 times, consisting of 5 sets of 6 measurements. Horizontally positioned spherical electrodes with a diameter of 12.5 mm are used for these measurements. The electrode gap is 2.5 mm and 1.0 mm for AC and LI breakdown voltage measurements, respectively. The rate of rise of the voltage is set as 2 kV/s in AC breakdown voltage measurements. The up-and-down method procedure is followed to be able to measure LI breakdown voltages [16]. These breakdown voltage measurements and the experimental procedures before/after the measurements are detailed in the authors' previous work [13].

III. RESULTS AND DISCUSSIONS

The main characteristics of AC and LI breakdown voltage measurements of natural ester and fullerene nanofluids are discussed in Table 1.

In AC breakdown voltage measurements, the mean and standard deviation data of the samples are calculated. The

average AC breakdown voltage of nanofluids with 0.05 g/L, 0.1 g/L and 0.2 g/L fullerene concentrations is lower than the base fluid. This characteristic change can be observed in low-concentration nanoparticle-added natural ester-based nanofluids. This reduction rate was observed around 15% in natural ester-based nanofluids using nanoparticles such as Fe₃O₄, Al₂O₃ and SiO₂ [4, 17]. The AC breakdown voltage strength of nanofluids added with 0.3 g/L and 0.4 g/L C₆₀ increased by 5.1% and 7.8%, respectively, compared to natural esters. The increase in breakdown voltage with C₆₀ concentration can be explained by the nanoparticle's strong electronegativity, photon absorption ability and high static dielectric constant [11, 12].

The LI breakdown voltage performance of nanofluid added with 0.1 g/L C₆₀ is 8.2% better than natural ester, see Table 1. Similarly, the LI breakdown voltage of the samples with 0.05 g/L, 0.2 g/L and 0.3 g/L concentrations are also better than the natural ester. This performance decreases dramatically in nanofluid added with 0.4 g/L C₆₀. The increase in LI breakdown voltage in nanofluids with lower concentration of nanoparticles can be explained by electron

TABLE I. MAIN CHARACTERISTICS OF AC AND LI BDVs [13]

		NE	0.05 g/L	0.1 g/L	0.2 g/L	0.3 g/L	0.4 g/L
AC BDV	Mean (kV)	66.7	58.3	62.6	63.2	70.1	71.9
	Std. Dev. (σ)	5.9	3.3	4.1	4.0	5.0	5.6
	Increment (%)		-12.6	-6.1	-5.2	5.1	7.8
LI BDV	Mean (kV)	75.7	78.9	81.9	79.7	78.0	67.3
	Std. Dev. (σ)	5.3	4.3	5.9	5.6	6.9	3.5
	Increment (%)		4.2	8.2	5.3	3.3	-11.1
	BDV Time (μ s)	3.39	1.75	2.82	2.34	1.61	2.44
	Velocity (km/s)	0.29	0.58	0.35	0.43	0.62	0.41

TABLE II. ANDERSON-DARLING TEST OF CONFORMITY TO WEIBULL DISTRIBUTION OF AC BDVs

	W	p-value	Conformity of Weibull Distribution
NE	0.5449	0.1611	Accept
0.05 g/L	0.2381	0.7620	Accept
0.1 g/L	0.3660	0.4361	Accept
0.2 g/L	0.3114	0.5328	Accept
0.3 g/L	0.2018	0.8676	Accept
0.4 g/L	0.2934	0.5784	Accept

TABLE III. ANDERSON-DARLING TEST OF CONFORMITY TO WEIBULL DISTRIBUTION OF LI BDVs OF NE AND NFs

	W	p-value	Conformity of Weibull Distribution
NE	0.3512	0.4716	Accept
0.05 g/L	0.6760	0.0698	Accept
0.1 g/L	0.5982	0.1099	Accept
0.2 g/L	0.5769	0.1334	Accept
0.3 g/L	0.4078	0.3485	Accept
0.4 g/L	0.4374	0.2964	Accept

trapping and tunneling mechanisms [18]. The superior electrical properties of C_{60} nanoparticles capture and slow down the electrons between the electrodes as the streamer develops [11]. In nanofluid samples with more than 0.2 g/L C_{60} concentration, the percolation threshold is exceeded and C_{60} nanoparticles form a conductive layer with the tunneling mechanism [11, 12]. The dramatic decrease in LI breakdown voltage performance after samples with 0.2 g/L C_{60} concentration can be explained by exceeding the percolation threshold.

The velocity of LI breakdown voltages of nanofluids is higher than that of natural ester. The increase in this velocity in low-concentration nanofluids can be explained by the formation of a more energetic streamer mechanism with increasing trap charge in fullerene nanofluids [12, 13]. The main reason for the increase in velocity in highly concentrated nanofluids is the development of the tunneling mechanism as a result of the decreasing distance between the nanoparticles and the faster movement of electrons in the streamer channel [11, 13].

In order to calculate withstand voltages in different breakdown voltages of AC and LI breakdown voltages, the conformity of these measurements with statistical distribution should be tested. The hypothesis of whether the distribution of these measurements fits the 5% significance level ($\alpha=0.05$) is determined using the Anderson-Darling test. The Anderson-Darling normality test is very sensitive to the distribution of data outside the mean and is a frequently used method for measurement data [3, 4].

W and p -value parameters are used to test the statistical distribution conformity hypothesis of this test; W being the coefficient of concordance and the p -value is defined as the probability of error in the statistical law compliance test of the measurement data. In the Anderson-Darling test, W must be lower than 1.5786 in order for the hypothesis to be accepted in the 0.05 significance level [19]. The hypothesis is accepted when this p -value is greater than the confidence interval 0.05 [3]. According to Table 2 and Table 3, both W and p -value meet the limit values defined for the hypothesis to be accepted by the Anderson-Darling test. These results show that both AC and LI breakdown voltage measurements fit the statistical distribution for all samples and withstand voltages can be calculated at different breakdown voltage probabilities.

AC withstand voltages at 1%, 10% and 50% breakdown voltage probabilities are shown in Table 4. The best performance in all AC breakdown voltage probabilities is observed in nanofluid added to 0.4 g/L C_{60} . The withstand voltages of all nanofluid samples are better than natural ester at 1% AC breakdown voltage probability. This performance increase of 0.3 g/L and 0.4 g/L C_{60} added nanofluids is quite remarkable as 16.0% and 22.3%, respectively. Compared to natural ester, the lowest withstand voltage performance in all breakdown voltage possibilities is 0.05 g/L C_{60} added nanofluid. The increase in withstand voltage performance of nanofluids at low breakdown voltage possibilities is due to the fact that the standard deviation of natural ester's breakdown voltage measurements is larger than nanofluids, see Table 1. Since the large standard deviation indicates that the measurements are distributed over a wider voltage range, the withstand voltage limit is reduced at a 1% breakdown voltage probability.

TABLE III. AC WITHSTAND VOLTAGES AT DIFFERENT BREAKDOWN PROBABILITIES DEDUCED FROM WEIBULL DISTRIBUTION

BDV Probability (%)	NE AC BDV (kV)	Concentration of NFs	AC BDV (kV)	Increment (%)
1	47.32	0.05 g/L	48.14	1.7
		0.1 g/L	50.46	6.6
		0.2 g/L	52.84	11.7
		0.3 g/L	54.88	16.0
		0.4 g/L	57.88	22.3
10	57.46	0.05 g/L	53.90	-6.2
		0.1 g/L	56.95	-0.9
		0.2 g/L	58.50	1.8
		0.3 g/L	63.10	9.8
		0.4 g/L	65.37	13.8
50	67.95	0.05 g/L	59.07	-13.07
		0.1 g/L	62.80	-7.6
		0.2 g/L	63.47	-6.6
		0.3 g/L	70.61	3.9
		0.4 g/L	72.35	6.5

TABLE V. LI WITHSTAND VOLTAGES AT DIFFERENT BREAKDOWN PROBABILITIES DEDUCED FROM WEIBULL DISTRIBUTION

BDV Probability (%)	NE LI BDV (kV)	Concentration of NFs	LI BDV (kV)	Increment (%)
1	60.68	0.05 g/L	64.82	6.8
		0.1 g/L	63.83	5.2
		0.2 g/L	61.80	1.8
		0.3 g/L	60.58	-0.2
		0.4 g/L	56.72	-6.5
10	68.50	0.05 g/L	72.60	6.0
		0.1 g/L	73.64	7.5
		0.2 g/L	71.69	5.0
		0.3 g/L	69.50	1.5
		0.4 g/L	62.17	-9.2
50	76.56	0.05 g/L	79.49	3.8
		0.1 g/L	82.62	7.9
		0.2 g/L	80.78	5.5
		0.3 g/L	78.57	2.6
		0.4 g/L	66.81	-12.7

The withstand voltages of natural esters and nanofluids in 1%, 10% and 50% breakdown voltage probabilities of LI breakdown voltage measurements are given in Table 5. At 1% breakdown voltage probability, the best LI withstand voltage performance compared to natural ester is 6.8% in 0.05 g/L C_{60} added nanofluid. The performance of nanofluid added with 0.1 g/L C_{60} is 5.2% better than natural ester at this puncture stress probability. The LI withstand voltages of the nanofluid added with 0.1 g/L C_{60} at 10% and 50% breakdown voltage probabilities are 7.5% and 7.9% better, respectively, than the base fluid. The worst LI withstand voltage performance in all rupture voltage possibilities is observed in the nanofluid with 0.4 g/L C_{60} .

The withstand voltages at 1% and 10% breakdown voltage possibilities are an important parameter in determining the

safety limits in the design and operation of power system equipment [20]. These withstand voltages show a remarkable improvement over natural ester in both AC and LI breakdown voltage possibilities.

IV. CONCLUSIONS

The main findings of this study, which investigated the AC and LI breakdown voltage characteristics of natural ester-based C₆₀ nanofluids, are as follows,

- AC breakdown voltages of nanofluids added with 0.3 g/L and 0.4 g/L C₆₀ are 5.1% and 7.8%, respectively, compared to natural ester.
- LI breakdown voltage performance of nanofluids is better than natural esters in all samples except 0.4 g/L C₆₀ added nanofluid. The best LI breakdown voltage performance is in 0.1 g/L C₆₀ added nanofluid with 5.9% increase.
- At 1% breakdown voltage probability, the AC withstand voltage performance of nanofluids added to 0.3 g/L and 0.4 g/L C₆₀ is 16.0% and 22.3% better, respectively, than natural ester. All nanofluid samples outperform natural ester at withstand voltages at this breakdown voltage probability.
- 0.05 g/L and 0.1 g/L C₆₀ added nanofluids have better LI withstand voltages than natural ester in all breakdown voltage possibilities. This difference is 68% and 52%, respectively, at 1% breakdown voltage probability.
- The LI breakdown voltage velocities of nanofluids are higher than that of natural esters. It is thought that the main reason for this phenomenon is the trap charge in low-concentration nanofluids and the tunneling mechanism in high-concentration nanofluids.
- The AC and LI breakdown voltage measurements and withstand voltages of nanofluids outperform natural ester, revealing their potential to be used in high voltage equipment such as power transformers.

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