Primo, V. A., Garcia, B., Burgos, J. C., & Perez, D. (2020). AC breakdown voltage of Fe3O4 based nanodielectric fluids. Part 1: Analysis of dry fluids. *In IEEE Transactions on Dielectrics and Electrical Insulation, 27*(2), 352–359

DOI:10.1109/tdei.2019.008277

AC Breakdown Voltage of Fe_3O_4 based nanodielectric Fluids. Part 1: Analysis of dry fluids

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ABSTRACT

The interest in developing new nanodielectric fluids suitable for electrotechnical applications has increased significantly in the last decade. Several authors have reported experiences on fluids manufactured using different base fluids and nanoparticles. Most of these studies are focused on the comparison of the thermal and dielectric properties of the nanofluids and the base liquids and most authors have found that the addition of nanoparticles to insulating liquids can lead to an improvement of their dielectric properties. This two-parts paper analyzes the enhancement of the AC Breakdown Voltage of several nanodielectric fluids prepared by dispersing variable concentrations of Fe_3O_4 nanoparticles in a mineral oil. In the first part of the study, the performance of fluids that had been previously dried and degasified fluids were tested. In the second part of the study the impact of the presence of moisture in the dielectric strength of the liquids is analyzed trying to get insight into the physical mechanisms that justify the observed enhancements when NP are added to insulating liquids.

Index Terms —**Insulating liquids; nanoparticles; nanodielectric fluids; AC breakdown voltage; Fe_3O_4; Weibull distribution**

1 INTRODUCTION

POWER transformers are an essential part of electrical systems. Failures in a power transformers can cause serious disruptions in the distribution of electricity and severe economic losses. An essential part of power transformers is the electrical insulation which is generally composed of two different materials: The first of these is the solid insulation, usually composed of Kraft paper and pressboard. The second is an insulating fluid that fills the transformer tank and acts as an electrical insulator and refrigerator.

Generally, the liquid insulation of transformers is mineral oil (MO) which is a petroleum derivative that is modified in order to obtain adequate results of both electrical insulation and transformer coolant. In recent years insulating fluids of other origins, such as natural and synthetic esters, are starting to be used as well.

Recently, several authors have proposed the application of nanotechnology to the insulation of transformers. Although a few works are referred to solid insulation most authors attempt to obtaining insulating fluids of superior properties by adding Nanoparticles (NP) to mineral oils and ester-based fluids. Different studies [1]–[3] show how the addition of conductive, semiconductive or insulating NP, with diameters in the range 10 to 60 nm, enhance the thermal and the dielectric properties of these oils [4]. These fluids, generally named dielectric nanofluids (NDF).

One of the first authors to work in this field was Segal, who studied the thermal and dielectric properties of colloidal fluids formed by dispersed NP in a mineral oil matrix and this author came to measure an improvement of up to 42% in AC Breakdown Voltage (AC BV) of the oil with nanoparticles [5], [6]. In addition to studies of AC BV, studies of Impulse Breakdown have been carried out obtaining great improvements as in [1]–[3], [7].

Several explanations have been given to the superior dielectric performance of NDF compared that of the base fluids; some of these are related with the effect that that the NP could have on the behaviour of the moisture molecules that are present on the liquids. [1]–[3]

In this two-parts paper the AC BV of several Fe_3O_4 based NDF is tested, investigating the evolution of this parameter with the moisture content of the fluids. In the first part paper, the manufacturing process and testing procedure are described and the AC BV of the MO and the different NDF fluids are analysed for dry conditions. The statistical analysis of the results is carried out using parametric and non-parametric analysis. The obtained results are compared with those published by other authors, and the fluids' performance is discussed in the light of different theories. In the second part of this work [8], the same fluids were subjected different conditioning processes in order to obtain NDF with different moisture contents and the evolution of the AC BV with the moisture content of the liquids was studied. A theoretical analysis is included for these aspects as well.

2 PREPARATION OF THE NANODIELECTRIC FLUIDS

The experimental study presented in this work is based on the evaluation of the AC BV of several NDF based in a commercial MO and a Fe_3O_4 NP dispersion. The materials used to prepare the NDF under test, the manufacturing process and the testing procedure are described below.

2.1 PREPARATION OF THE NDF

A commercial dispersion of Fe_3O_4 NP of diameter 10 nm suspended in a mixture of hydrocarbons, silicon compounds and non-flammable oils up to a concentration 50 % was used to produce the NDF under test. The NP dispersion was manufactured by MAGRON (South Korea), and commercialized by the company Supermagnete (Germany) under the name MFR-DP1. In this type of products the NP are coated with surfactants to inhibit aggregation; this fact has an impact on the stability of the obtained liquids [9].

Small volumes of this liquid were dispersed in the commercial MO Nytro 4000X (Nynas AB, Sweden) to obtain samples of NDF with NP concentrations 0.05 g/L, 0.1 g/L, 0.2 g/L. Fluids that remained stable at ambient temperature for more than ten months [9] were obtained following the procedure described below.

Firstly, the amount of NP suspension that must be added to the base fluid was calculated taking into consideration the total amount of NDF to be prepared, the required NP concentration and the percentage of NP in the MFR-DP1 fluid.

Then the calculated masses of MFR-DP1 fluid were added to beakers that contained 0.5 L of Nynas Nytro 4000X. The mixture were performed using an sonicator with an ultrasonic probe with wave intensity 268 W/cm2. Each mixture was stirred for two hours in intervals of 30 seconds of agitation and 30 seconds of pause, to avoid overheating of the fluid. A photography of the obtained fluids is shown in Fig. 1.

Figure 1: Oil samples with different concentrations of NP. From left to right: 0 g / L, 0.05 g / L, 0.1 g / L and 0.2 g / L.

After ultrasonic stirring, the samples were kept at ambient temperature for 24 hours to allow the release of the bubbles generated by the sonicator, and finally the samples were placed in a vacuum oven where they were dried and degasified at 50 ºC and 0.1 atm for 48h.

2.2AC BREAKDOWN VOLTAGE TESTS

One of the main factors to determinate the insulating properties of a dielectric oil is the AC BV i.e. the minimum voltage level at 50 Hz that causes the dielectric breakdown of the oil. In this work AC BV measurements of the prepared NDF were carried out according to IEC60156 standard [10] using a tester Baur DTA 100C. The configuration of electrodes considered in this work was sphere-sphere with a gap distance 2.5 mm. The rate of voltage increase was 10 kV/s until breakdown, as indicated by IEC60156 standard [10], and the test frequency was 50 Hz

For each AC BV measure, the following measuring sequence was followed:

1. The test cell is filled with 500 mL of the fluid under analysis
2. A sample of the fluid under test is taken to obtain its initial moisture content with Karl Fischer method, and then the sample is left in magnetic stirring for five minutes to release any air bubbles that have been formed during the filling of the test cell.
3. Ten measurements of AC BV with Baur DTA 100C are carried out. Between each rupture the fluid is left two minutes under magnetic stirring.
4. A new sample of fluid is taken for moisture determination and ten additional breakdowns are applied.
5. The sequence 1 to 4 is repeated for a fresh sample of fluid.

For each considered condition 40 AC BV values were recorded (i.e. 20 breakdown measurements on two different samples) and the moisture content of the samples was measured four times during the tests.

3 RESULTS

Figure 2 shows the 40 AC BV values measured on the NDF prepared with NP concentrations 0.05 g/L, 0.1 g/L, 0.2 g/L and the AC BV measured on samples of MO. As explained before, the 20 first points and the next 20 points correspond to different samples of fluid. It is important to note that the maximum voltage that could be applied with the deployed tester was 98 kV, so some of the measurements appear truncated. This happens specially for the NDF with concentration 0.2 g/L.

As can be seen, the measurements do not follow any tendency in relation to the measuring sequence but they have a significant dispersion. A statistical analysis of the results has been carried out following either a parametric and a non-parametric approach.

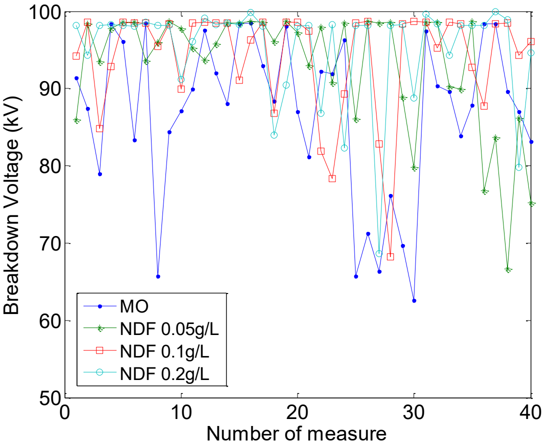

Figure 2. AC BV measurements in the fluids under test

3.1 STATISTICAL ANALYSIS

Firstly, a non-parametric statistical analysis of the AC BV measurements is carried out. Table I presents the values of the mean, the median and the standard deviation of the AC BV values measured for each analysed fluid. In addition, the values of percentiles 1, 2, 50 and 90 are displayed.

TABLE I. Statistical analysis of the AC BV measurements

AC BV	MO	NDF 0.05 g/L	NDF 0.1 g/L	NDF 0.2 g/L
Mean (kV)	87.02	93.14	94.12	95.21
Median (kV)	89.00	96.65	98.40	98.20
Std dv	10.38	7.74	6.92	6.59
Percentile 2 (kV)	63.53	69.25	71.23	71.96
Percent. 50 (kV)	89.00	96.65	98.40	98.20
Percent. 90 (kV)	98.40	98.70	98.60	99.00

Several conclusions can be drawn from these calculations. In the first place the values of both the mean and the median of the AC BV rise as the concentration of NP in the NDF is increased. Improvements of a 10 % are observed for the NDF with NP concentration 0.2 g/L. Moreover, the standard deviation of the measurements decreases as the concentration of NP becomes higher. This can be clearly observed in Figure 3., which shows the histograms of the measurements obtained on all the analysed fluids. As can be seen, as the concentration of NP rises, the values of the AC BV are more concentrated around 98 kV, which is the maximum voltage that can be applied by the applied tester. This means that most of the samples did not broke at 98 kV, so the real breakdown voltages of the NDF would be even higher than those displayed in Table I.

On the other hand, the study of the percentiles, shows that the lower breakdown voltages registered on the NDF increase significantly compared with those recorded in the MO (i.e. percentile 2 is 63.53 kV for MO and 71.96 kV for the NDF with concentration 0.2 g/L. This parameter is important for the transformer design.

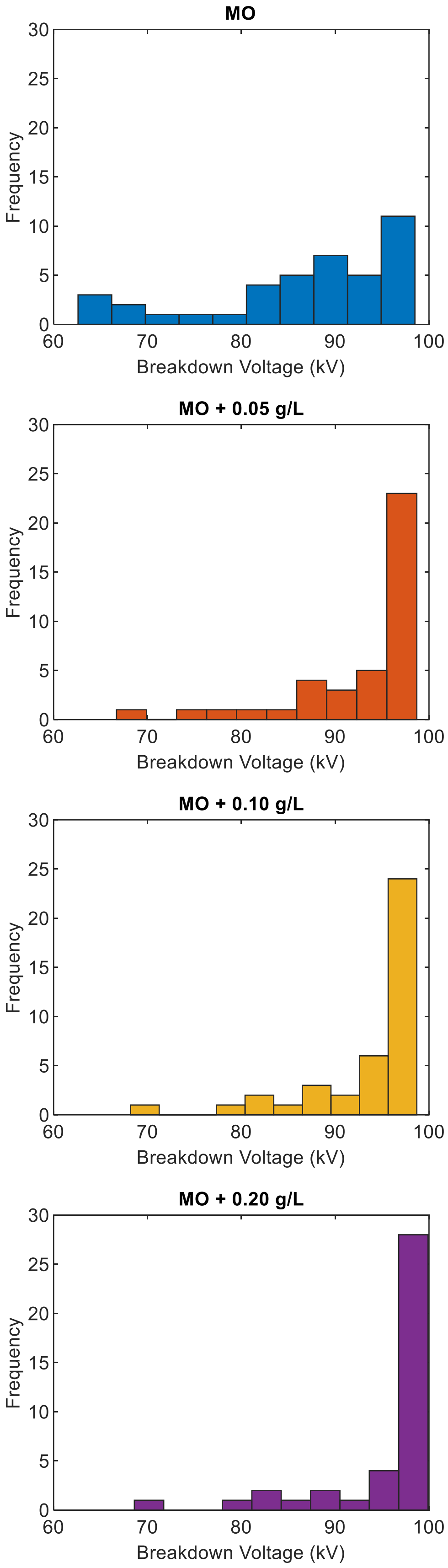

Figure 3: Frequency of ACBV for each fluid at 10% RS a) MO, b) MO + 0.05 g/L NP c) MO + 0.1 g/L NP d) MO + 0.2 g/L NP

In order to analyze the performance of NDF when higher masses of Fe_3O_4 NP are added, fluids with NP concentrations 0.3 g/L and 0.4 g/L were prepared and tested. It must be said that the long-term stability of these liquids is poorer than those observed for lower NP concentrations. Fig 4 shows the average value of the AC BV of the fluids with NP concentrations within 0 and 0.4 g/L. As can be seen for concentrations above 0.2 g/L the AC BV drops, although the values recorded for these NDF are still better than those of MO.

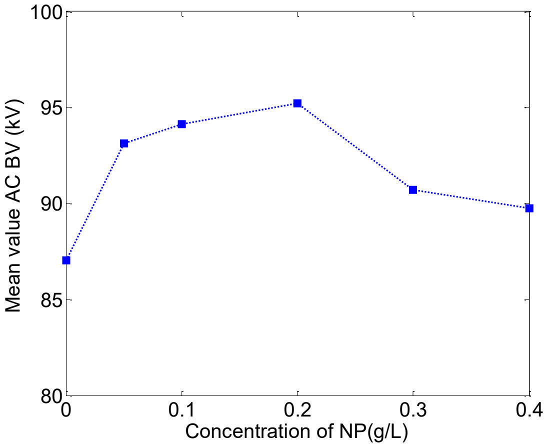

Figure 4: Mean value of the AC BV vs. concentration of Fe_3O_4 NP

3.2 WEIBULL ANALYSIS

To complete the previous study, the measurements were analyzed according to Weibull distribution. For that analysis, the fluids with concentrations 0.05, 0.1 and 0.02 g/L were considered, as they were the ones with better performance either from the dielectric as from the stability point of view.

The 40 values of ACBV obtained for each liquid were adjusted to a Weibull distribution (eq (1))

$$P\,(V) = 1 - e^{-\left(\frac{V}{a}\right)^{b}} \qquad (1)$$

where P(V) is the failure rate of the test liquid at voltage V, a is the scale parameter, which predicts the AC BV at a failure probability of 63%, and *b* is the shape parameter, which gives a measure of the scattering of the measurements.

Table II shows the values of parameter *a* and *b* in the Weibull adjustment and Fig. 5 shows the cumulative probability of failure vs the AC BV for the different fluids. As can be seen, the addition of NP leads to an improvement of the dielectric strength, especially for the case of the NDF prepared with concentration of NP 0.2 g/L.

TABLE II. PARAMETERS FOR THE WEIBULL ADJUSMENT OF THE AC BV OF THE DIFFERENT FLUIDS

FLUID	*a*	*B*
MO	86.0	19.6
NDF 0.05 g/L	90.6	15.9
NDF 0.10 g/L	92.8	16.2
NDF 0.20 g/L	95.1	16.4

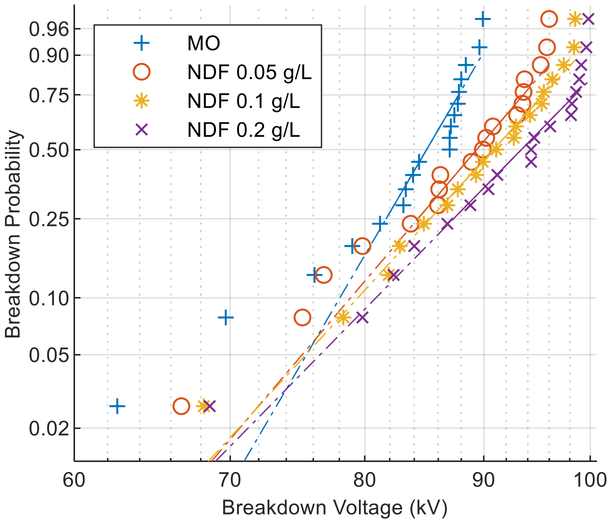

Figure 5: Fitting of the data to a Weibull distribution.

Table III provides the AC BV estimated with the Weibull distribution for the MO and the different NDF for different probabilities of failure. As can be seen, the Weibull analysis estimates improvements up to 15 % for the NDF at high probabilities of failure. The accuracy of the estimations corresponding to low probabilities is not good enough because, as other authors have described [10], [11], the adjustment of the data to the fitting curve in this zone is not adequate.

TABLE III. BREAKDOWN VOLTAGE FOR DIFFERENT FAILURE PROBABILITIES

	AC Breakdown Voltage (kV)			
Failure Probability	**MO**	**NDF 0.05 g/L**	**NDF 0.1 g/L**	**NDF 0.2 g/L**
5%	75.5	75.2	75.5	76.5
10%	78	78.8	79.5	80.2
25%	81.5	84.3	85.2	87.1
50%	85.0	89.4	90.8	93.4
75%	87.8	93.6	95.6	98.6

4 DISCUSSION

The experimental measurements shown in this work demonstrate that the addition of Fe_3O_4 NP in low concentrations achieve significant improvements in the AC BV values, reduces the dispersion of the measurements and increases the breakdown voltages at low probabilities of failure, what is important for the transformer's insulation design. This improvement might seem surprising if we consider that the NP are conductive and that in fact, the resistivity of the resulting NDF fluids are higher that of the MO (Figure 6)

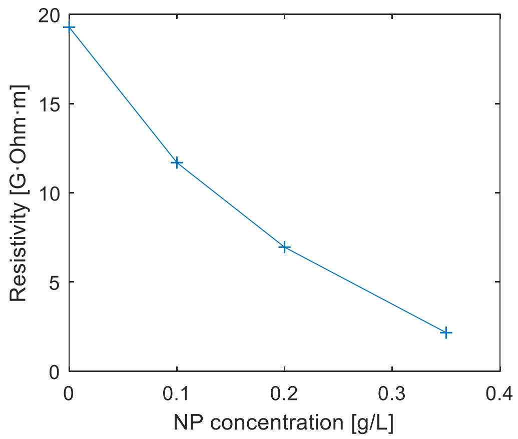

Figure 6: Resistivity of the fluids under analysis at different NP concentrations.

The increase of the breakdown voltage, has been generally explained by the Electrical Double Layer (EDL), according to which when a NDF is subjected to an electric field, the positive ionic charges accumulate on the surface of the NPs, attracting negative ionic charges around them [13], [14]. Thus, an electrical double layer (EDL) is formed. The volume of the EDL close to the NP surfaces is called the compact layer, and consists of immobile negative ions attracted strongly to the NP surfaces. The net charge density in the compact layer drops gradually with increasing distance from the NP surfaces, reaching zero in the electrically neutral area of the fluid. The ions in this region, called the diffuse layer of the EDL, are less affected by the electrostatic interaction with the NP and thus have higher mobility [15]. The formation of the EDL in a NDF subjected to an electric field is probably the main reason for the improvement in dielectric properties reported in this work and by some other authors [15], [16].

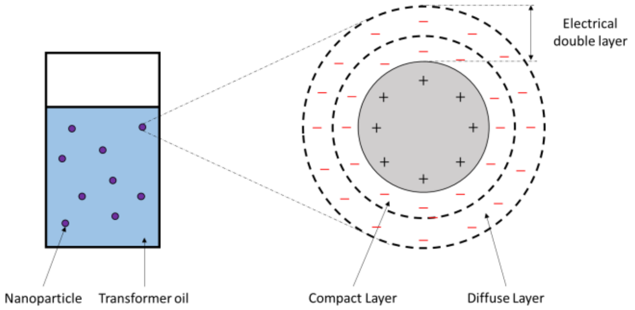

Figure 1. Electrical double layer formation in NDF. Adapted from [13], [15].

Hwang [17] proposed that the effect of dispersing a certain type of NP in a base fluid on the dielectric breakdown process depends on the NP relaxation time, which is defined as:

$$\tau = \frac{2\varepsilon_{BF}+\varepsilon_{NP}}{2\sigma_{BF}+\sigma_{NP}} \qquad (2)$$

where ε_{BF} and ε_{NP} are the permittivity of the base fluid and the NP and σ_{BF} and σ_{NP} are their conductivities.

If the relaxation time of the NP is shorter than the time scale of the electrodynamic process being considered, the effect of the NP on the process will be very noticeable, while if the relaxation time is longer than the time scale of the process the NP will have little effect on the breakdown process.

Considering the resistivity of the MO used in this work, and the typical values of the relative permittivity of MO and Fe_3O_4 NP reported in the literature, the relaxation times of the prepared NF was calculated obtaining times of $7.47\cdot10^{-14}$ s (for MO). These times are much smaller than the time scale of the breakdown processes, and thus the observed improvement would be justified in the light of Hwang's theory.

Some works present experimental results that cannot be justified with the electron-scavenging model and the relaxation time constant [18] . Du et al [19] propose an alternative explanation for the observed improvement, based in the increase of the shallow trap density of dielectric fluids when NP are present. The authors demonstrate that repeated electron trapping and de-trapping processes could be one of the main charge transport processes in NDF; if the fast electrons, generated in presence of an intense electric field are trapped and de-trapped by the shallow traps when moving from high to low field locations, their speed, and so the speed of the streamer propagation, will drop significantly. The described mechanism explains the enhancement of the dielectric properties observed in NDF based in semi-conductive and insulating NP and probably has a role in the results obtained in this work as well.

5 CONCLUSIONS

In this work, the AC BV of several NDF, prepared by dispersing Fe_3O_4 NP in a commercial MO has been tested. In this first part of the paper the tests are carried out on dry fluids' samples while in the second part of this paper the performance of these fluids in high moisture environments is tested. The behavior in relation to moisture has been cited as a differential factor of NDF by some authors.

The analysis of the measurements demonstrates that the AC BV of the MO can be significantly enhanced by adding low concentration of NP to it. As shown in this work, all the analyzed parameters in relation to the breakdown process improved: the mean value of the breakdown voltage, the breakdown voltage at low failure probabilities and the standard distribution of the measurements. These improvements are in line with those reported by other authors and can be explained in the light of the electron-scavenging model and by the increase of the shallow trap density in the NDF.

Although, these results suggest that the addition of NP could improve the reliability of the MO from the point of view of the dielectric strength, many other issues must be solved before those materials can be safely applied to real transformers.

ACKNOWLEDGMENT

This work has been supported by the Spanish Ministry of Economy and Competitiveness within the project DPI2015-71219-C2-2-R.

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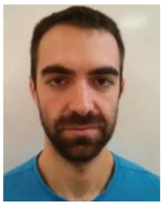
BIOGRAPHIES



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