Streamer simulation in nano-based dielectric fluids at different Fe_3O_4 nanoparticle concentrations

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Abstract Nano-based dielectric fluids (NDF) seem to be a good alternative for improving dielectric and thermal characteristics of conventional liquid dielectric systems used in power transformers. Fe_3O_4 (magnetite) nanoparticles (NP) is one of the most investigated type of NP. Some experiments have shown that with its addition to mineral oil (MO) and ester, increases in breakdown voltage (BV) can be achieved applying both AC and DC voltages. This kind of NP have the advantage that can be handled and synthesized safely and easily using a two-steps method. Besides, streamer propagation can be reduced or avoided introducing Fe_3O_4 NP in dielectric fluids. These NP act as electronic traps and behave like slow-moving charged particles in the NDF. This behaviour as well as other dielectric characteristics such as resistivity, permittivity and loss tangent, depend on the type, size and concentration of NP. In this work, comparisons and analyses of thermal and dielectric performance of NDF with different Fe_3O_4 NP concentration are made taking into account the evolution and behaviour of streamers. It has been found that temperature in streamer tip, its length and speed depend on the Fe_3O_4 NP concentration and the BV is affected because of changes in streamer speed. The most adequate concentration for controlling the streamer has been obtained through simulations and comparisons with experimental results showing good agreement.

I. INTRODUCTION

Nano-based dielectric fluids (NDF) are used for improving thermal and electrical characteristics of conventional liquid dielectric systems such as mineral oil (MO) and ester. Additionally, NDF can improve the power density and reliability in power transformers and other important equipment for the power systems. Nanoparticles (NP) immersed in dielectric fluids can produce a reduction on the charge density inside the NDF and create a lower and uniform internal electric field, which indeed gives a reduction on the partial discharges (PD) occurrence. The addition of conductive particles increases the thermal conductivity even at low concentrations [1].

Depending on its conductivity, NP may be classified into three groups [1], [2], conducting (Fe_3O_4 , Fe_2O_3 , ZnO , SiC),

semiconducting (TiO_2 , CuO , Cu_2O) and insulating (Al_2O_3 , SiO_2 , BN). Although, Fe_3O_4 is classified as conducting, some researchers using Raman spectroscopy, X-Ray diffraction, UV-Vis spectroscopy and Scanning Electron Microscopy have characterized it as semiconductive [3].

Fe_3O_4 (magnetite) is one of the most investigated type of NP for NDF applications because some experiments have shown that with the incorporation of magnetite NP in transformer oil have given better dielectric strengths in comparison with the base fluid. In the case of magnetite NDF with mineral oil as base fluid, it could result in increases in the AC breakdown voltage (ACBV) up to 42.8% [4] and for DC breakdown voltage (DCBV) up to 21.4% [5]. With positive lightning impulses, breakdown voltages of magnetite NDF showed improvement for a 10% w/v concentration [6], [7]. In addition, magnetite NP inside NDF act as electronic traps and behave like slow-moving charged particles in the NDF, which reduces both, the streamer velocity and streamer length propagation [8]. This behaviour as well as other dielectric characteristics such as resistivity, permittivity and loss tangent, depend on the type, size and concentration of NP [3].

Furthermore, magnetite NP could be used with ester fluids, that have higher biodegradability and fire resistance compared with traditional mineral oil [9], with an increase in ACSV of 19.8% [10]. Magnetite NP has applications from bio-medical; as a contrasting agent for magnetic resonance imaging and therapeutic agent for cancer treatment, to space as high-performance seals component [11], and as NDF with propylene carbonate, is used in high-performance lithium batteries [12]. This kind of NP have as advantage to be handled and synthesized safely and easily using a two-steps method where the NP are previously prepared by chemical or physical methods and then dispersed in the base fluid using an adequate mixing process for bringing good stability and viscosity at different types of concentrations even for diverse moisture content [1], [2]. However, NDF with magnetite are ferrofluids and external magnetic fields can cause some agglomerations that affect the dielectric performance [5]. When there is moisture, NDF have similar dielectric properties than base fluid, however, more research in this subject is required [13].

In this paper, comparisons and analysis of thermal and dielectric performance of NDF with different Fe_3O_4 NP concentration are made considering the evolution and behaviour of streamers. A model of positive streamer process in NDF, developed in COMSOL Multiphysics and considering the IEC 60897 [14] configuration, is used to perform Finite-Element Method (FEM) simulations. Length, speed and temperature of streamer have been calculated when fast rise step voltages have been applied. Additionally, the quantification of different NDF parameters required for streamer simulations; such as NP radius, NDF electric permittivity and the surfactant effect are studied and experimental values, reported by other authors, are considered.

The paper is organized as follows: first, the streamer simulation parameters for NDF with different NP concentrations are defined and its determination procedures are reviewed in section II, then parameters calculations and results of streamer propagation are presented in section III. Finally, some conclusions are drawn in section IV.

II. STREAMER SIMULATION IN NDF

Streamer behaviour analysis is important because it allows the understanding of the pre-breakdown phenomena in liquid insulating materials [15]. Besides, positive streamers are of main interest in comparison with negative ones, this is because they propagate faster than negatives, resulting in lower breakdown voltages during shorter periods of time [16], [17].

In this study, a mathematical model implemented in COMSOL Multiphysics and presented by the authors in [17] is modified in order to analyse different NP concentrations as well as the effect of surfactant. The performance of NDF under overvoltages is mainly dependent on the electric charge dynamics in NP [8], which is related to NP attachment time constant, (1), and the upper limit of electric charge density in NP for trapping electrons, (2).

$$\tau_{pc} = 4 \epsilon_{UTR} / (\mu_e \rho_e) \quad (1)$$

$$\rho_{np,sat} = Q_s / (V_{NP} / \phi_{NP}) \quad (2)$$

In (1) and (2): τ_{pc} (s) is the NP time constant; ϵ_{UTR} (F/m) is the mineral oil electric permittivity; μ_e ($\text{m}^2\text{V}^{-1}\text{s}^{-1}$) is the mobility of electrons; ρ_e (C/m^3) is the volumetric charge density of electrons; $\rho_{np,sat}$ (C/m^3) is the upper limit of electric charge density in NP for trapping electrons, Q_s (C) is saturation charge; V_{NP} (m^3) is NP volume; and ϕ_{NP} is the volume magnetic fraction of magnetite NP in NDF.

A. Nanoparticles Size

From (1) and (2) can be observed that in order to simulate the NDF, geometrical and electromagnetic parameters have to be determined for each specific concentration under analysis. The determination of NP size is a hard task and results depend on the experimental procedure. Table I summarizes some values of magnetite NP diameter measured using different experiments. All experiments referred in Table I were made in NDF with magnetite NP using UTR-40 mineral oil as base fluid. As can be seen, the value of NP diameter is dependent on the experimental procedure, exhibiting different values for the same sample and concentration.

The stability of NDF is verified using the zeta potential measurements since the measured quantity indicates the degree of electrostatic repulsion between similarly charged adjacent particles.

TABLE I. MAGNETITE NP MEASURED DIAMETER THROUGH DIFFERENT EXPERIMENTAL PROCEDURES

Report	Measured NP Diameter		
	Diameter	Experiment	Ref.
1	14 nm	X-Ray Diffraction	[18]
2	10 – 30 nm	Scanning Electron Microscopy	
3	8.12 nm	Magneto-Granulometric Analysis	
4	31.4 nm	Zeta Potential	[19]
5	28.65 nm	X-Ray Diffraction and Spectroscopy	[20]

However, Jarial and Rao [19] also used this test for determining the diameter of magnetite NP in the NDF. Furthermore, the radius of NP can be determined using measurements of magnetization and magnetic saturation.

In [20], using X-ray diffraction and spectroscopy, the following magnetic characteristics of magnetite were found: χ_i (initial susceptibility) = 0.12278, $\mu_0 M_s$ (saturation magnetization density) = 0.0063 T, giving an average NP diameter of 28.65 nm. Finally, in the study made by Pîslaru et al. [18] using magneto-granulometric analysis, it was found that the average magnetic diameter of the magnetite particles was 6.46 nm with a standard deviation of 2.18 nm and a physical average diameter of 8.12 nm. Furthermore, considering a surfactant layer of 1.9 nm in thickness, the hydrodynamic diameter was 11.92 nm.

B. Electric Permittivity and Effect of Surfactant

When analysing NDF, at least three different elements have to be specified; the base fluid, the NP material and surfactant. In this study, the base fluid is mineral oil, the material of NP is magnetite and the surfactant is oleic acid. Regarding the relative permittivity of UTR-40 mineral oil, this is a well-known parameter and its value is between 2.1 and 2.2 [8], [18].

Surfactants prevent agglomeration of particles caused by the Van der Waals forces, giving a stable NDF. Oleic acid is the most common substance used as surfactant [2]. It is almost constant with frequency [21], but depends inversely on temperature [22]. Typical values for resistivity, loss tangent and relative permittivity in NDF with oleic acid as surfactant are, respectively, $2.05 \times 10^9 \Omega\text{m}$, 0.360 and 2.35 [7]. Timko et al. [23] made tests in NDF with UTR-40 mineral oil and magnetite NP with oleic acid as surfactant in different NP concentration and found that the relative permittivity increases with the volume of concentration from 2.23 for 1.62% to 2.7 for 3.03%.

For determining the electric permittivity of NDF, the Maxwell-Garnett equation can be used [18]:

$$\epsilon_{NDF} = \epsilon_{UTR} + 3(a/b) \quad (3)$$

Where ϵ_{UTR} [F/m] is the electric permittivity of mineral oil, $a = \phi_{NP} \epsilon_{UTR} (\epsilon_{NP} - \epsilon_{UTR})$, $b = \epsilon_{NP} + 2 \epsilon_{UTR} - \phi_{NP} (\epsilon_{NP} - \epsilon_{UTR})$, ϵ_{NP} is the electric permittivity of magnetite NP.

Surfactants, such as oleic acid, are long chain molecules with an atomic structure in which one termination is hydrophobic and the other is hydrophilic [24]. In this way, it allows steric isolation between the NP associated with magnetic and Van der Waals attractions and at the same time, NP with surfactant act as moisture traps, which promotes the creation of a water layer around NP causing a slight increase

in the relative permittivity of the NP themselves. In addition, around the NP and the water layer, an intermediate transition region between the oil and the previous set (specifically the water layer) must be considered, which is known as the interface or interaction zone [25], [26]. Considering that NP and water have a very similar relative permittivity, Negri [27] proposed to consider an equivalent NP of radius equal to the radius of the NP plus the thickness of the water layer and with an equivalent permittivity equal to the NP one. A hydrodynamic diameter of 11.92 nm is assumed taking into account the NP diameter and the surfactant and water layer.

III. RESULTS

NP concentration affects the dielectric strength of synthesized NDF. The first concentration considered is 1.62 % w/v, taken from Timko et al. [23]. This value is used because its experimental reported values allow calculating the dynamic charge parameters shown in Table II, which are required for simulating streamer propagation process. Three additional concentrations were assumed corresponding to 10%, 50% and 75% of the volume magnetic fraction of magnetite NP in the NDF corresponding to 1.62% w/v NP concentration. Values presented in Table II were calculated using (1)–(3) and with the following assumptions: $\epsilon_{UTR} = 2.2\epsilon_0$ F/m; $\epsilon_{NP} = 81\epsilon_0$ F/m; $\mu_e = 1e-4$ m²V⁻¹s⁻¹; $\rho_e = -1000$ C/m³; $Q_s = -2.61e-18$ C; $\tau_{pc} = 7.79e-10$ s. In order to consider highly conductive NP, such as magnetite, Hwang [8] proposed a limit corresponding to 80% of the upper value of electric charge density in NP for trapping electrons as calculated with (2) and a NP time constant equal to 2 ns. For implementing the simulations, the electrodes configuration established by the IEC 60897 standard [14] with a gap of 25 mm and a step voltage of 300 kV in magnitude with a rise time of 10 ns was applied. Fig. 1 shows the electric field distribution along the z-axis (streamer in length, $L_{streamer}$) in the needle-sphere electrodes configuration at 1000 ns for concentrations presented in Table II and the results for mineral oil (0% w/v of NP) are superposed for comparison.

Fig. 2 shows temperature distribution along z-axis in the needle-sphere electrodes configuration at 1000 ns for concentrations presented in Table II and in the UTR-40 mineral oil. Table III summarizes the results for the considered concentrations and the maximum value of temperature along the symmetry axis. As can be seen, the addition of magnetite NP reduces both, streamer propagation velocity and streamer length, from a value of 89.688% for a concentration of 0.162%, to 95.253% for 1.62%w/v comparing with the result obtained for mineral oil without NP (UTR-40). Besides, the time-to-breakdown is increased from 869.762% for a concentration 0.162% to 1190.820% for 1.62% w/v compared with the result for the base fluid.

TABLE II. ELECTRIC CHARGE PARAMETERS FOR SIMULATING DIFFERENT NDF CONCENTRATIONS

Variable	NP concentration (%)			
	0.162	0.81	1.215	1.62
$\mu_0 M_s$ (T)	3.493e-4	1.747e-3	2.620e-3	3.493e-3
ϕ_{NP}	6.238e-4	3.119e-3	4.679e-3	6.238e-3
ϵ_{NDF} (F/m)	1.957e-11	1.992e-11	2.014e-11	2.037e-11
$\rho_{np,sat}$ (C/m ³)	-1.835e+3	-9.175e+3	-1.376e+4	-1.835e+4

Fig. 1. Electric field distribution along the needle-sphere z-axis at 1000 ns for concentrations under 1.62 % w/v NP.

Fig. 2. Electric field distribution along the needle-sphere z-axis at 1000 ns for concentrations under 1.62 % w/v NP.

TABLE III. STREAMER SIMULATION RESULTS FOR DIFFERENT NP CONCENTRATIONS

Variable	NP concentration (%)				
	0 (UTR-40)	0.162	0.81	1.215	1.62
$ E_{max} $ (V/m)	3.875e+8	3.483e+8	3.508e+8	3.784e+8	4.413e+8
$L_{streamer}$ (mm)	1.1385	0.1174	0.1175	0.1081	0.0882
Streamer propagation velocity (km/s)	1.1385	0.1174	0.1175	0.1081	0.0882
Time-to-breakdown (μ s)	22.310	216.354	216.170	234.968	287.982
T_{max} (K)	790.765	358.828	355.478	367.747	317.825

Finally, the maximum temperature along the z-axis exhibits a reduction from 54.623 % for 0.162 % concentration to 59.808 % for 1.62% w/v.

In Table IV, it is shown several results for streamers when it is applied lightning impulses, 1.2-microsecond rise, to mineral oil and NDF. These experiments were made using needle-sphere electrodes configuration at 25.4 mm, with positive impulses and Fe₃O₄ NP. The highest increase in the time-to-breakdown, 116.667%, and the lowest average streamer velocity, -53.774%, were reported by Segal et al. [4] however the specific concentration is not detailed.

TABLE IV. COMPARISON AMONG STREAMERS RESULTS.

Fluid	Concentration	Time-to-break-down [μs]	Variation [%]	Average streamer velocity [km/s]	Variation [%]	Ref.
Univolt 60 M	0	12	0	2.12	0	[4]
Univolt-Colloid NDF	Non Specified	26	116.667	0.98	-53.774	
Nytro 10X M	0	16	0	1.59	0	
Nytro-Colloid NDF	Non Specified	25	56.250	1.02	-35.849	

On the other hand, Wang et al. [6] for a concentration of 10% w/v reported an increase in the time-to-breakdown of 12.279% and a reduction on the average streamer velocity of 10.995%. As it is depicted, the variation among the results presented in [4, 6, 7] and those obtained by simulation in this document are due to the applied voltage is a 300 kV pulse with a rise time of 10 ns in a gap of 25 mm.

IV. CONCLUSIONS

NDF can reduce the streamer propagation velocity compared with the one in mineral oil without NP. It can be achieved by including the adequate volume concentration of NP in the base fluid. As example, a 1.62 % w/v concentration of magnetite type NP can reduce the streamer length from 1.14 mm for mineral oil to 0.09 mm when NP are added. Besides, the temperature can be reduced from 790.8 K to 317.8 K under the same scenarios. An appropriate volume concentration of NP immersed in the base fluid can be obtained by simulating the streamer propagation with finite-element software. According with the results shown for the cases studied the effect in decreasing temperature and BV is reduced with lower NP concentrations. Finally, additional simulations non-presented in this work have depicted that increasing the concentration up to 3 % w/v do not improve the benefit of reducing thermal and streamer length compared with those obtained for 1.62 % w/v.

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