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Article

Streamer Inception from Ultra-Sharp Needles in Mineral Oil Based Nanofluids

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Abstract: Positive and negative streamer inception voltages from ultra-sharp needle tips (with tip radii below 0.5 μm) are measured in TiO_2 , SiO_2 , Al_2O_3 , ZnO and C_{60} nanofluids. The experiments are performed at several concentrations of nanoparticles dispersed in mineral oil. It is found that nanoparticles influence positive and negative streamers in different ways. TiO_2 , SiO_2 and Al_2O_3 nanoparticles increase the positive streamer inception voltage only, whilst ZnO and C_{60} nanoparticles augment the streamer inception voltages in both polarities. Using these results, the main hypotheses explaining the improvement in the dielectric strength of the host oil due to the presence of nanoparticles are analyzed. It is found that the water adsorption hypothesis of nanoparticles is consistent with the increments in the reported positive streamer inception voltages. It is also shown that the hypothesis of nanoparticles reducing the electron velocity by hopping transport mechanisms fails to explain the results obtained for negative streamers. Finally, the hypothesis of nanoparticles attaching electrons according to their charging characteristics is found to be consistent with the results hereby presented on negative streamers.

Keywords: streamer inception; electric discharges; nanofluids; mineral oil

1. Introduction

A couple of decades ago, it was experimentally observed that some nanoparticles (NPs) improve the dielectric strength of host dielectric liquids [1]. This finding was counterintuitive to common knowledge suggesting that particles decrease the dielectric strength of any host liquid [1]. Nevertheless, subsequent experiments have confirmed the dielectric strength improvement due to the presence of some NPs added into dielectric liquids. Some overall facts have been deduced from these experimental studies. It has been concluded that smaller NPs yields better effects than larger NPs, that there is an optimal concentration of NPs, and that their effect depends on the characteristics of the liquid and the NPs [1–4].

The dielectric strength of a dielectric liquid is characterized by its electric breakdown withstand capability. In most of the literature, the dielectric strength of liquids is studied following standards such as IEC 60897 (e.g., [5–8]). However, breakdown in dielectric liquids is a highly complicated, not well understood process, involving electronic, thermal and mechanical phenomena taking place within fractions of microseconds [9–11]. For this reason, only few numerical models of pre-breakdown phenomena can be found in the literature (e.g., [12–14]), which unfortunately cannot fully explain all experimental findings [15]. Without a clear understanding of the breakdown phenomena in base oils, it is difficult to interpret and explain the effect of NPs on the breakdown phenomena. In spite of this, three main hypotheses have been postulated in the literature explaining the dielectric strength improvements of hydrocarbon oil due to the presence of NPs. They can be summarized as: (1) water

adsorption on the NPs surface inhibits detrimental effects of moisture on dielectric strength [16], (2) NPs attach electrons converting them into slower ions [5], and (3) increments of electronic trapping and de-trapping processes due to NPs reduce the electron velocity [6].

Electric breakdown in hydrocarbon oils is the consequence of the development of so-called streamers. Hence, the study of streamers and other pre-breakdown phenomena is key to understand the effect of NPs on the electric breakdown process. It is generally accepted that negative streamers in base oils initiate from electronic avalanches in the liquid phase followed by light emission, a shock wave and the formation of a gas bubble [9–11]. The mechanism of initiation of positive streamers is nevertheless less understood [17], although it has been observed to be influenced by moisture content [18].

In general, the streamer inception has been studied for needles with tip radii larger than $0.5\ \mu\text{m}$ (e.g., [17,19]). However, it is generally difficult to theoretically interpret these experimental results since the processes of injection of charges into the liquid can only be quantified for sharper needles [17]. Considering that the self-consistent numerical simulation of injection of charges from ultra-sharp needles (with tip radii less than $0.5\ \mu\text{m}$) and its associated electrohydrodynamic liquid motion has been recently reported [20], the measurement of streamer initiation from ultra-sharp needles is the next step to enable the future physical analysis of this process using computer simulation. It also provides an interesting scenario to test whether the proposed hypotheses can explain the effect of NPs in host oil.

In this study, positive and negative streamer inception voltages of nanofluids prepared with five different kinds of NPs are investigated for ultra-sharp needles. Differences between positive and negative streamer inception voltages are highlighted and the existing hypotheses are discussed based on the reported results. The paper is organized as follows. Section 2 introduces the experimental conditions of this work. Section 3 presents the results of the experiments. The main hypothesis explaining effects of NPs are discussed in Section 4. Finally, conclusions are presented at the end of the paper.

2. Experimental Details

2.1. Electrode Configuration

Streamers are initiated in a needle-sphere electrode configuration. Since ultra-sharp needles are made using electro-etching of 1 mm diameter tungsten wires, their tip radius R_{tip} range between 110 nm and 350 nm. The tip of the needles is separated by a distance d_{gap} to stainless steel spheres with diameter of 10 mm. The electrode configuration is placed within a 16 mm inner diameter container with borosilicate cylindrical glass walls, filled with the liquid under test. The experimental setup is shown in Figure 1. Since the used ultra-sharp needles are very fragile, they are often damaged by strong discharges occurring mainly at positive polarity or by handling accidents. For this reason, several needles are used throughout the experiments as reported in Table 1.

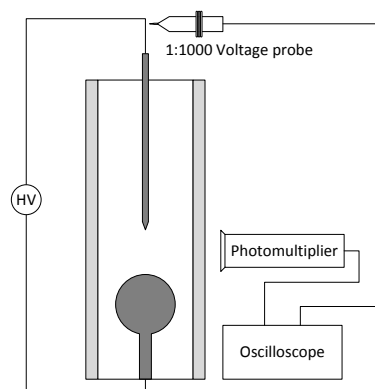


Figure 1. Experimental set up is shown. Streamers are initiated in a needle to sphere electrode configuration. A photomultiplier is used to detect the light emitted by the streamer. Both light and voltage signals are recorded with an oscilloscope.

Table 1. Nanofluids preparation details and results of the streamer inception measurements.

NF Preparation			Electrode Configuration	Weibull Parameters			Streamer Inception Voltage and Laplacian Electric Field			
NP	wt %	Cluster Size (nm)		η	β	γ	$V_{50\%}$ (V)	E_{LAP} (MV/cm)	% Change	
ZnO Size: 20 nm Hydrophobic $\tau < 0.1$ ns $\rho_m = 5.6$ g/cm ³	0	-	Negative Streamer $d_{gap} = 4.5$ mm $R_{tip} = 225$ nm	2403	5.85	3439	5696	30.8	-	
	0.01	150		5588	8.43	1761	7111	38.4	24.8%	
	0.02	165		3760	4.77	3534	7016	37.9	23.2%	
	0.05	380		5758	7.17	1686	7157	38.7	25.7%	
	0	-	Positive Streamer $d_{gap} = 3$ mm $R_{tip} = 200$ nm	1265	3.92	4516	5668	33.4	-	
	0.01	150		1183	3.37	5271	6331	37.3	11.7%	
	0.02	165		1700	4.00	5051	6602	38.9	16.5%	
	0.05	380		1168	2.62	5727	6743	39.8	19.0%	
	SiO ₂ 10–20 nm Hydrophilic $\tau \sim 30$ s $\rho_m = 2.6$ g/cm ³	0	-	Negative Streamer $d_{gap} = 1.3$ mm $R_{tip} = 350$ nm	1981	4.25	3752	5569	21.8	-
0.001		-	2595		5.55	2874	5303	20.8	-4.8%	
0.002		-	1972		4.68	3733	5557	21.8	-0.2%	
0.005		-	1800		3.83	3782	5417	21.2	-2.7%	
0.01		-	1568		3.12	4077	5472	21.4	-1.7%	
0.05		-	2387	5.46	3281	5512	21.6	-1.0%		
0		-	Positive Streamer $d_{gap} = 1.5$ mm $R_{tip} = 170$ nm	1955	4.02	3130	4915	36.5	-	
0.001		-		2400	4.91	2598	4824	35.8	-1.8%	
0.002		-		1080	2.91	3684	4636	34.4	-5.7%	
0.005		-		1290	2.37	3764	4869	36.1	-0.9%	
0.01		-		2650	4.90	3253	5711	42.4	16.2%	
0.05		-		2985	1.53	4835	7184	53.3	46.2%	
Al ₂ O ₃ 20 nm Hydrophilic $\tau \sim 40$ s $\rho_m = 4$ g/cm ³		0		-	Negative Streamer $d_{gap} = 1.3$ mm $R_{tip} = 350$ nm	1294	2.92	4248	5390	21.1
	0.005	270		1697		3.61	4041	5575	21.8	3.4%
	0.01	230	1895	3.75		3853	5571	21.8	3.4%	
	0.02	325	1875	3.08		4061	5726	22.4	6.2%	
	0.05	310	1858	3.90		3870	5561	21.8	3.2%	
	0	-	Positive Streamer $d_{gap} = 1.5$ mm $R_{tip} = 170$ nm	1349	1.97	4032	5152	38.2	-	
	0.005	270		1854	4.18	3763	5461	40.5	6.0%	
	0.01	230		2376	2.53	3762	5817	43.1	12.9%	
	TiO ₂ 20 nm Hydrophilic $\tau \sim 25$ s $\rho_m = 3.8$ g/cm ³	0		-	Negative Streamer $d_{gap} = 1.38$ mm $R_{tip} = 350$ nm	1085	1.92	4696	5592	21.6
0.001		270	1451	3.09		4286	5574	21.5	-0.3%	
0.002		255	972	1.97		4775	5582	21.6	-0.2%	
0.005		250	1904	4.12	3756	5497	21.3	-1.7%		
0		-	Positive Streamer $d_{gap} = 4.65$ mm $R_{tip} = 150$ nm	1916	2.48	4097	5750	37.3	-	
0.001	270	2192		1.77	4730	6512	42.2	13.2%		
0.002	255	3679		4.84	4804	8214	53.3	42.9%		
0.005	250	2476		2.39	5834	7958	51.6	38.4%		
C ₆₀ 0.7 nm Hydrophilic 1 ps $< \tau < 50$ s $\rho_m = 1.65$ g/cm ³	0	-	Negative Streamer $d_{gap} = 1.63$ mm $R_{tip} = 350$ nm	1284	2.96	4725	5860	21.9	-	
	0.002	-		1666	3.38	4550	6045	22.5	3.2%	
	0.005	-		1893	3.47	4566	6269	23.4	7.0%	
	0.01	-		2500	3.94	4493	6771	25.3	15.5%	
	0	-		Positive Streamer $d_{gap} = 1.13$ mm $R_{tip} = 300$ nm	2894	8.29	2206	4975	25.66	-
	0.002	-	1366		4.06	3913	5161	26.62	3.7%	
	0.005	-	1243		3.12	4706	5812	30	16.8%	
	0.01	-	3577		1.96	4609	7576	39.1	52.3%	

2.2. Preparation of Nanofluids

Nanofluids are prepared by adding dried NPs with different mass fractions into filtered and degassed Nytro 10XN mineral oil. The mineral oil has a mass density of 880 kg/m³ and a relative electric permittivity of 2.2. In order to discuss the electron attachment hypothesis [5], the set of NPs chosen have associated relaxation time constants lower and higher than the microsecond time scales of the streamer development. Similarly, the NPs chosen range from highly hydrophilic to highly hydrophobic in order to discuss the water absorption hypothesis [16]. Five different NPs are used: laboratory-grade zinc oxide (ZnO-C₁₈) silane-coated NPs [21] and commercial-grade NPs of aluminum

oxide (Al_2O_3), titanium oxide (TiO_2), silicon oxide (SiO_2) and fullerene (buckminsterfullerene C_{60}) distributed by IoLiTec. The size of the NPs is equal to or lower than 20 nm, which is expected to lead to a stronger effect in the nanofluid compared with larger NPs (e.g., [2]). Their respective relaxation time constants, hydrophilicity, mass density ρ_m and further parameters are reported on Table 1. Since the additives and surfactants generally used to treat the surface of NPs may influence the physical and dielectric properties of nanofluids (e.g., [22]), they are not used here during the preparation of the nanofluids. In this way, changes in the streamer inception voltage of the prepared nanofluids can be attributed exclusively to the properties of the NPs.

The NPs are dispersed in the fluid by using an ultrasonic bath for 30 min. Moisture content is not measured in these experiments, but an estimate of 10 ppm is expected from previous work of the authors (e.g., [23]). In order to synthesize the nanofluids at low NP concentration, a large amount of liquid is first prepared and only a fraction is used in the experiments. After preparation, a sample of the used nanofluid is taken immediately for size distribution analysis while the streamer inception tests are performed. The aggregate size distribution of the nanofluids are measured at 25 °C using dynamic light scattering (DLS) with a Malvern Zetasizer Nano ZS instrument. Even though nanofluids tend to agglomerate with time (e.g., [24]), the DLS measurements have shown that the used nanofluids are stable throughout the duration of the test. However, observe that the long-term stability of the used nanofluids has not been measured since it is outside the scope of this paper. The measured aggregates average sizes (AS) of the used NPs are also reported in Table 1.

2.3. Streamer Testing Procedure

The inception of streamers is measured under a voltage ramp V_R obtained by charging a capacitor. The voltage V_R follows the equation:

$$V_R = 12 \text{ kV} \left[1 - \exp\left(-\frac{t}{3.2 \text{ } \mu\text{s}}\right) \right]. \quad (1)$$

The duration of the voltage ramp is 5 μs . Positive or negative streamers are incepted by applying the voltage ramp to the needle or to the sphere electrode, while the opposite electrode is connected to the ground.

It should be noted that the shape of the applied voltage differs from traditional voltages waveforms reported in the literature, such as AC 50 Hz [25], DC waveforms [26], lightning impulse [27] or rectangular waveforms [28]. The main reason for choosing a fast voltage ramp lies on the stability of the nanofluids. Nanoparticles dispersed on mineral oil can rapidly agglomerate and sediment, affecting the electrical behavior of the nanofluid. Therefore, the complete test duration should be as short as possible to avoid changes in the nanofluids properties. Since each applied voltage ramp provides an inception voltage value suitable for direct statistical sample analysis, sufficient datapoints can be generated within a short time. Observe that other voltage waveforms would require a significantly larger number of experiments in order to reach the same accuracy level since they require multi-level test methods.

The inception of streamers is identified by detecting their emission of light with a Hamamatsu R11540 photomultiplier placed 100 mm away from the electrodes. The experiments are performed under dark conditions in order to avoid any stray light reaching the photomultiplier. The streamer inception voltage V_{inc} is defined as the instantaneous voltage at the moment when the first light signal pulse is detected. A typical measurement of the applied voltage V_R , light signal and estimated V_{inc} are shown in Figure 2.

The streamer inception voltage determined with this light detection method has been compared with that obtained using a charge measurement technique similar to that developed in [17]. Although both techniques lead to similar results (not shown here), the light detection technique is more sensitive since it can detect streamers with an electric charge lower than 0.05 pC. Therefore, the light detection technique is here preferred over a charge measurement due to its sensitivity, simplicity and reliability.

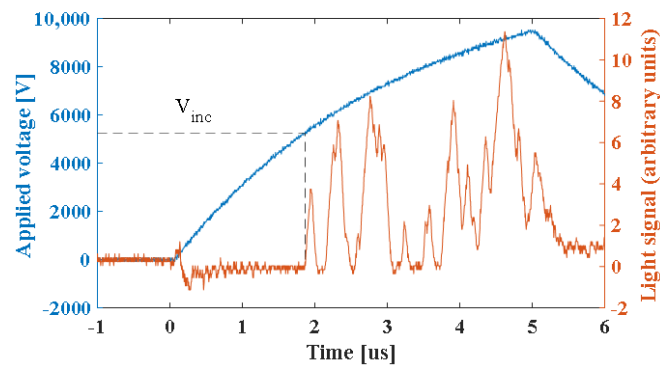


Figure 2. Typical applied voltage V_R and light signal recorded as a function of time. The streamer inception voltage V_{inc} is also indicated.

For each nanofluid under each polarity, 200 voltage ramps are applied with a waiting time of three seconds in between them. V_{inc} is estimated for each voltage ramp, and used to build the empirical cumulative distribution function (CDF) of the test. The three-parameter Weibull function [29] used to fit the data follows the equation:

$$F(V_{inc}) = 1 - \exp\left(-\left(\frac{V_{inc} - \gamma}{\eta}\right)^\beta\right), V_{inc} > \gamma, \quad (2)$$

where γ , η and β are the location, scale and shape parameters of the Weibull function, respectively. Table 1 shows the parameters in all tests estimated using the Weibull++ software. The streamer inception experiments for each nanofluid under each polarity are first performed in mineral oil in the absence of any NP. In this manner, the reference CDF of the base liquid is obtained. Then, nanofluids with different concentrations are prepared and immediately tested.

Since streamers (especially those of positive polarity) lead to a breakdown of the liquid soon after their inception is reached, an insulating solid barrier is generally used to electrically isolate one electrode. This prevents streamers from bridging the entire gap and damage the electrodes [30]. Two reasons prevented the use of an insulating solid in the experiments hereby reported. The first is that the insulating solid accumulates charge over repeated experiments leading to a reduction of the electric field and ultimately increasing the streamer inception voltage. In order to prevent charge accumulation, sufficiently long waiting times between experiments must be provided in order to allow the charges to dissipate. However, experiments with nanofluids require a short duration of the test in order to avoid agglomeration of the nanofluid as already mentioned. The second reason is the possible interaction of NPs with the solid interface, which introduces a further uncertainty that is difficult to assess.

In order to reduce the possibility of breakdown, a large gap distance is initially set for each needle-sphere configuration. The gap distance is then reduced until the 50% probability streamer inception voltage $V_{50\%}$ of the CDF in mineral oil reaches values about half of the maximum applied voltage, at about 5 kV. This $V_{50\%}$ in mineral oil is chosen in order to be able to measure increments of the $V_{50\%}$ larger than 70% when nanofluids are tested under same electrode configuration. The gap distances set in the experiment are reported in Table 1.

The next step in the experiment is to measure the effect of the NPs on the $V_{50\%}$ relative to that of the base oil. For this reason, the CDF obtained for each nanofluid is compared to its reference CDF previously measured in mineral oil under the same electrode configuration. In this manner, the effect of each kind of NP relative to mineral oil can be addressed despite of the different electrode configurations set throughout the experiment. Since the $V_{50\%}$ of mineral oil is measured in all electrode configurations, it is also possible to perform a comparison of the Laplacian electric fields at the measured $V_{50\%}$ between the different electrode configurations used. Results of this analysis are reported in Section 4.1.

3. Streamer Inception in Nanofluids

3.1. Silicon Oxide, Titanium Oxide and Aluminum Oxide Nanofluids

Figure 3 shows the streamer inception probability curves obtained for the SiO₂ nanofluids under both polarities and the corresponding reference with mineral oil. The three-parameter Weibull fitting curves for each dataset are depicted as solid lines whilst the data points are depicted with markers. Figure 4 shows changes in the $V_{50\%}$ of SiO₂ nanofluids relative to the mineral oil. Error bars correspond to the 95% confidence bounds of the estimated $V_{50\%}$. No significant change on the negative streamer inception voltage is found within the tested concentrations in comparison with mineral oil. Likewise, no differences in the empirical distribution functions of the nanofluids and mineral oil under positive polarity are observed below a concentration threshold at about 0.005 wt %. However, SiO₂ NPs are found to have a significant impact on the $V_{50\%}$ of positive streamers above this threshold, as shown in Figure 4.

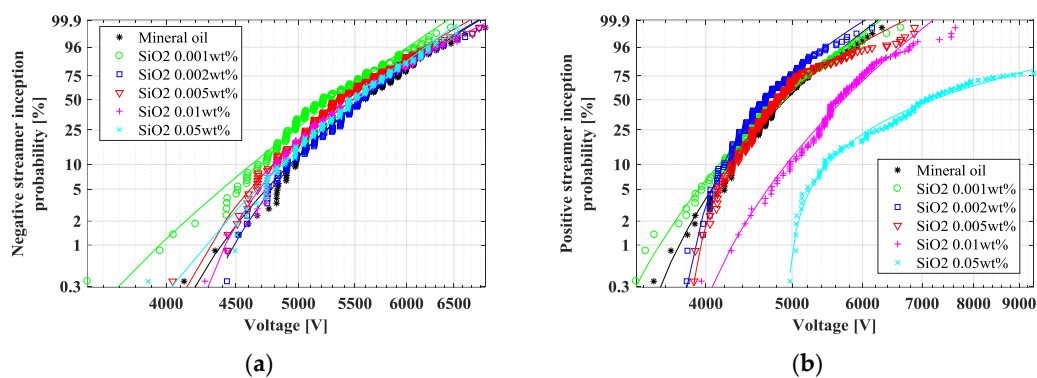


Figure 3. Streamer inception cumulative probability of SiO₂ nanofluids at different mass concentrations under (a) negative and (b) positive polarity. The reference data of mineral oil is added for sake of comparison. The corresponding fitting Weibull functions are shown as solid lines.

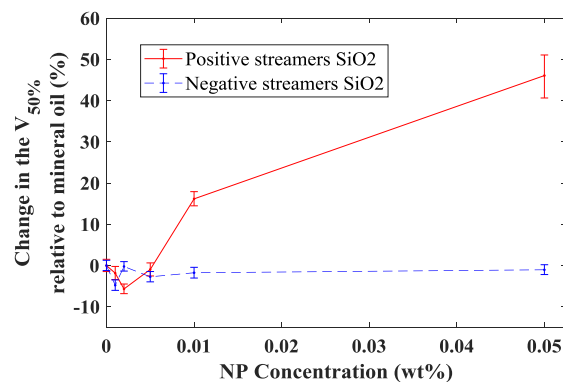


Figure 4. Change in $V_{50\%}$ of SiO₂ nanofluids relative to mineral oil as a function of mass fraction w_i .

The effect of TiO₂ nanofluids on the streamer inception voltages is similar to that obtained with SiO₂ nanofluids. Since there is no appreciable difference in the negative CFDs between mineral oil and TiO₂ nanofluids, they are not shown. The CFDs for positive streamers in TiO₂ nanofluids are shown in Figure 5 while the changes in $V_{50\%}$ relative to mineral oil are depicted in Figure 6. Observe that the $V_{50\%}$ of positive streamers increase with TiO₂ concentration to similar values than those of SiO₂ shown in Figure 4. However, the used concentrations of TiO₂ are one order of magnitude lower than for the SiO₂ nanofluid. No larger concentration of TiO₂ NPs is tested since the change in the $V_{50\%}$ of positive streamers already reaches a maximum level at the highest concentration used, as shown on Figure 6.

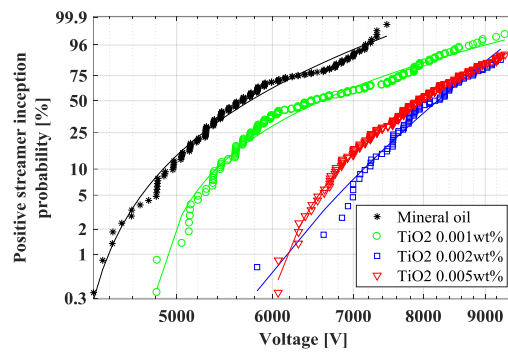


Figure 5. Streamer inception cumulative probability of TiO₂ nanofluids at different mass concentrations under positive polarity. The reference data of mineral oil is added for sake of comparison. The corresponding fitting Weibull functions are shown as solid lines.

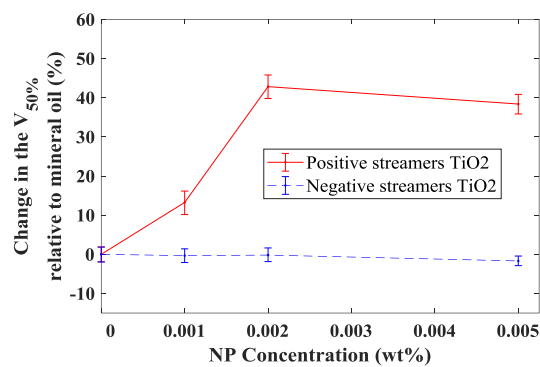


Figure 6. Change in $V_{50\%}$ of TiO₂ nanofluids relative to mineral oil as a function of mass fraction w_i .

Figure 7 depicts the $V_{50\%}$ change of the Al₂O₃ nanofluids relative to mineral oil for streamers of both polarities as a function of concentration. It is found that $V_{50\%}$ increases with concentration until a maximum peak value is reached at $w_i \sim 0.02$ wt %. A larger concentration yields in turn a lower $V_{50\%}$ relative to that at $w_i \sim 0.02$ wt %. Although the CFDs are not shown, the estimated three parameters of the Weibull function for the curves are reported in Table 1.

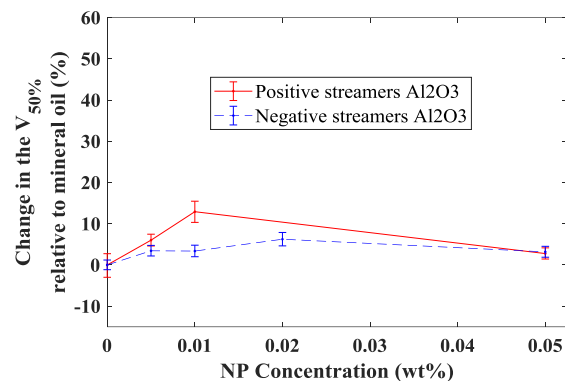


Figure 7. Change in $V_{50\%}$ of Al₂O₃ nanofluids relative to mineral oil as a function of mass fraction w_i .

In the literature, partial discharge (PD) experiments with SiO₂ nanofluids have been previously reported in a 20 mm gap needle-plane configuration under DC voltages [26]. It was observed that SiO₂ nanofluids increase the positive PD inception voltage relative to mineral oil, and at the same time, exhibit no effect on the inception voltage of negative PDs [26]. Even though the PD results measured

in [26] cannot be quantitatively compared with the $V_{50\%}$ here presented (since the experimental and physical conditions are different), they interestingly show the same consistent effect of SiO_2 nanofluids.

Streamer inception experiments with TiO_2 have also been recently reported in a needle-sphere electrode configuration with tip radius of 50–70 μm and 25 mm gap distance according to IEC 60897 [6]. It was observed in [6] that a TiO_2 nanofluid with concentration of 0.0227 wt % increases $V_{50\%}$ for positive streamers by 13.5% relative to mineral oil. Observe that the reported results for positive streamers also show an increase in $V_{50\%}$ when adding TiO_2 NPs. However, no direct comparison between the results in Figure 6 and in [6] can be done since they correspond to nanofluids with different concentrations and surface treatment procedures.

Breakdown voltage experiments with Al_2O_3 nanofluids have been previously reported in a needle-plane electrode configuration with gap distances lower than 0.3 mm [3]. It was observed that breakdown voltage of Al_2O_3 nanofluids at $w_i \approx 0.45$ wt % mass fraction concentration was lower than the breakdown voltage of the base oil, and that the breakdown voltage decreased as Al_2O_3 concentration increased [3]. This concentration is almost one order of magnitude larger than the highest concentration used in the experiments hereby reported for Al_2O_3 . Interestingly, the decreasing trend of $V_{50\%}$ under both polarities presented in Figure 7 for $w_i > 0.02$ wt % suggests that the concentrations used in [3] were already deleterious to the dielectric strength of mineral oil.

3.2. Zinc Oxide and C_{60} Nanofluids

Figures 8 and 9 show the streamer inception probabilities obtained for the ZnO and C_{60} nanofluids under both polarities and their corresponding reference with mineral oil. Observe that the $V_{50\%}$ of the nanofluids in both polarities is larger than that of the base fluid.

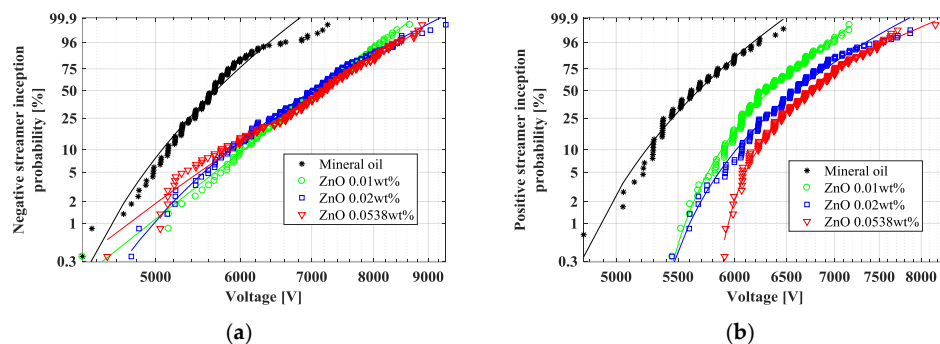


Figure 8. Streamer inception cumulative probability of ZnO nanofluids at different mass concentrations under (a) negative and (b) positive polarity. The reference data of mineral oil is added for sake of comparison. The corresponding fitting Weibull functions are shown as solid lines.

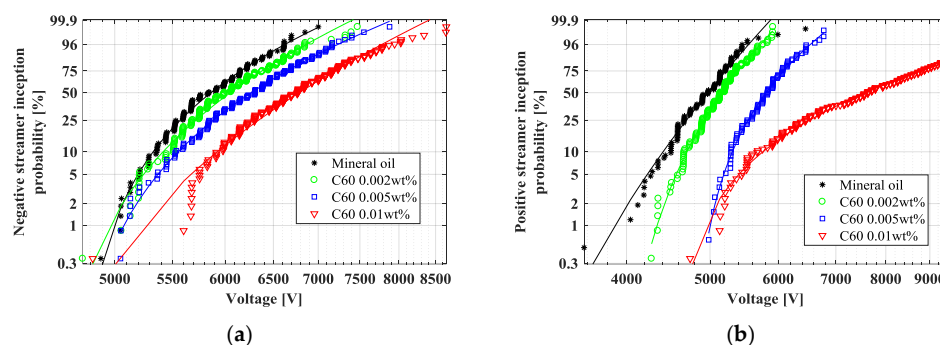


Figure 9. Streamer inception cumulative probability of C_{60} nanofluids at different mass concentrations under (a) negative and (b) positive polarity. The reference data of mineral oil is added for sake of comparison. The corresponding fitting Weibull functions are shown as solid lines.

The change of the $V_{50\%}$ of ZnO nanofluids relative to mineral oil as a function of NP concentration is depicted in Figure 10. As can be seen, $V_{50\%}$ of the nanofluid under negative polarity is independent of the NPs concentration within the tested range. Even though small differences in $V_{50\%}$ can be observed with concentration, they are attributed to the stochastic nature of the streamer inception process. On the other hand, the change on the $V_{50\%}$ under positive polarity increase with NP concentration.

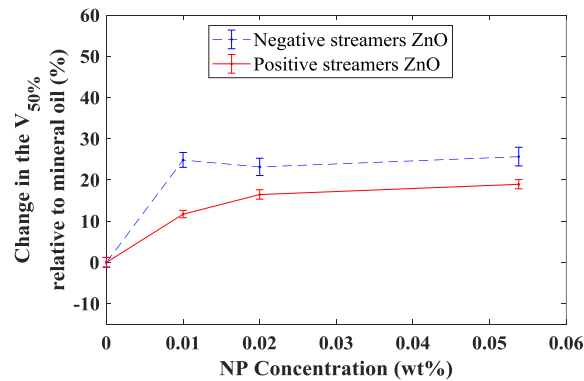


Figure 10. Change in $V_{50\%}$ of ZnO nanofluids relative to mineral oil as a function of mass fraction w_i .

The changes in $V_{50\%}$ of C_{60} nanofluids relative to mineral oil are depicted in Figure 11. $V_{50\%}$ of both polarities for C_{60} nanofluids increase with concentration. Interestingly, C_{60} is the only NP among those here studied that monotonically increases $V_{50\%}$ with concentration within the tested range. Concentrations of C_{60} are also lower than ZnO, Al_2O_3 and SiO_2 NPs, though yielding similar increments in the $V_{50\%}$. This might be explained by the results presented in [2], where it was experimentally found that the dielectric strength of nanofluids improves with reductions of NPs size. The dielectric strength in [2] was measured according to ASTM D-877 on nanofluids prepared at concentrations below 0.5 wt % with NPs sizes lower than 100 nm. In our case, Table 1 reports aggregates sizes lower than 1 nm for the tested C_{60} NPs in mineral oil, hence allowing these NPs to influence strongly the dielectric strength of the base oil at lower NP concentration.

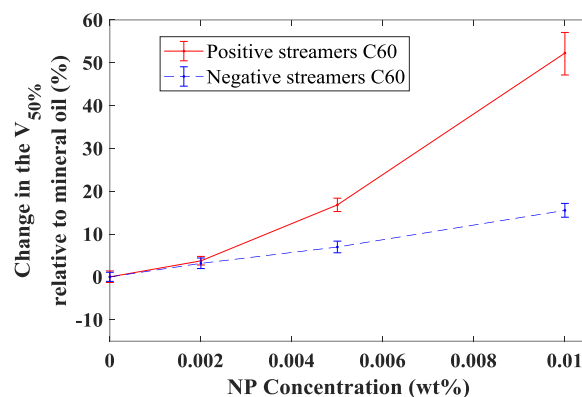


Figure 11. Change in $V_{50\%}$ of C_{60} nanofluids relative to mineral oil as a function of mass fraction w_i .

PD inception experiments in C_{60} nanofluids at concentration of 0.01 wt % presented in [26] reported increments in positive and negative inception voltages of about 14% and 12% relative to the base oil, respectively. Since the inception voltages for PDs reported in [26] are not equivalent to the $V_{50\%}$ here reported for streamers, they cannot be quantitatively compared. However, the results in [11] also show the consistent increase in the initiation voltage of prebreakdown phenomena in both polarities, as reported in Figure 11.

Breakdown voltages in ZnO nanofluids have been also previously reported [2,4]. These improvements in breakdown voltage compared to the reference are consistent with the increment of the $V_{50\%}$ of positive streamers relative to the base oil hereby presented.

4. Discussion

4.1. Streamer Inception in Mineral Oil

The evaluation of the streamer inception has been generally assessed in the literature based on the Laplacian electric field on the needle tip at $V_{50\%}$ [11,30]. In order to estimate the inception electric field E_{LAP} in the experiments in mineral oil reported in the previous section, the Laplace equation $\nabla^2 V = 0$ is solved using COMSOL Multiphysics for the different needle-sphere electrodes used in the experiment. Boundary conditions are set accordingly to the applied voltage on each streamer polarity. The calculations are proven to be mesh independent.

The Laplacian fields E_{LAP} for positive and negative streamers of the experiments performed in mineral are shown in Figure 12. It is found that E_{LAP} increases as R_{tip} decreases and that E_{LAP} of positive and negative streamers follow a similar trend. This finding is in good agreement with previously reported experiments in mineral oil with blunter needles [30]. However, the obtained values of E_{LAP} here reported are up to two times larger than those obtained by extrapolation of the results in [30]. A possible explanation for this behavior lies on the electrostatic screening of the actual surface electric field due to the strong generation of charged carriers from ultra-sharp needles [20]. Observe that electric charges under the high electric fields in front of ultra-sharp needle tips are produced due to field emission, field ionization, or similar charge generation mechanisms, as previously studied by the authors [20,23,31]. As the produced charges accumulate in front of the needle's tip under the applied voltage ramp, the surface electric fields are electrostatically shielded. Since there is currently a lack of knowledge on the effect of the space charge on the streamer inception process, further analysis of these experiments can be the subject of future simulation studies.

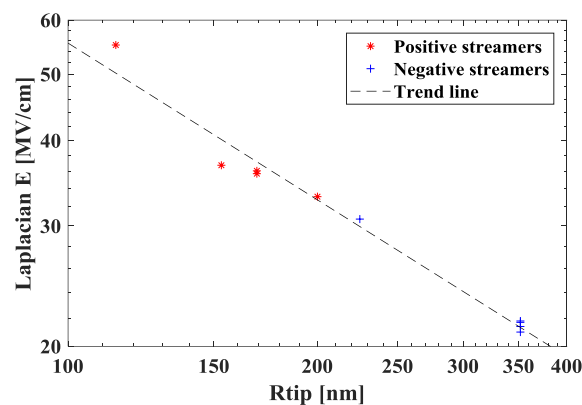


Figure 12. Laplacian electric field at the tip of the needles at $V_{50\%}$ for mineral oil as a function of R_{tip} .

4.2. Streamer Inception in Nanofluids

The initiation of streamers from the ultra-sharp needles in nanofluids also takes place under intense generation of charges, as discussed for mineral oil in the previous subsection. This intense production of charged carriers defines a special precondition for the inception of streamers. Even though the existing hypotheses of the dielectric effect of NPs [8–10] were proposed for blunter electrodes (where charge generation before streamer initiation is less important [30]), they should still apply under the experimental conditions here reported. Thus, these hypotheses are discussed in this section.

4.2.1. Water Adsorption Hypothesis

Increments in the positive partial discharge inception voltage reported in [26] were attributed to the moisture adsorption properties of the SiO₂ NPs. This hypothesis was further strengthened by the fact that hydrophobic silane coated SiO₂ NPs yields deleterious effects on dielectric strength, whilst untreated SiO₂ NPs improve the dielectric strength [32].

Interestingly, the water adsorption hypothesis is consistent with the results here presented for SiO₂ and TiO₂ nanofluids. Both nanofluids exhibit increments on the positive streamer inception voltage and a negligible effect on the negative streamer inception voltage relative to the base oil, as observed in Figures 4 and 6 and reported in [26]. Furthermore, TiO₂ exhibit superhydrophilicity when exposed to UV light [33]. The similar relative increments on $V_{50\%}$ of positive streamers for SiO₂ and TiO₂ nanofluids measured with concentrations of TiO₂ one order of magnitude lower than those of SiO₂ indicate a greater water uptake capability of the TiO₂ NPs, provided that the water adsorption hypothesis is the only mechanism for the improved positive streamer inception of TiO₂ nanofluids.

On the other hand, Al₂O₃ NPs are hydrophilic by nature, but chemical reactions with carboxylic acids can render their surfaces from highly hydrophilic to highly hydrophobic [34]. Although Nytro 10XN is a complex combination of hydrocarbons, its low acid number $AN < 0.01$ mg KOH/L suggests Al₂O₃ NPs dispersed in oil are still hydrophilic. In comparison to SiO₂ and TiO₂ nanofluids, the case with Al₂O₃ NPs show lower increments in the $V_{50\%}$ of positive streamers. This could be then explained if Al₂O₃ NPs dispersed in mineral oil are less hydrophilic than SiO₂ and TiO₂ NPs.

Furthermore, since C₆₀ and silane-coated ZnO NPs are hydrophobic [21,35], water adsorption cannot explain the increments in streamer inception voltages reported in Figures 10 and 11. These findings indicate that NPs influence the positive streamer inception process by other physical mechanism(s) in addition to water adsorption.

4.2.2. Shallow Traps Hypothesis

It was observed in [6] that there is an increment on the shallow trap density of TiO₂ nanofluids relative to the base oil. It was then hypothesized that TiO₂ NPs increased the electron trapping and de-trapping processes, reducing the electron velocity in nanofluids relative to the base oil. However, it is generally accepted that negative streamers in liquids are initiated by electronic processes in the liquid phase [10,11,36], which are influenced by the electron velocity and attachment processes. If TiO₂ NPs would reduce the electron velocity, an effective reduction of the $V_{50\%}$ for negative streamers would also be expected. However, results on TiO₂ here presented in Figure 6 show no changes in the $V_{50\%}$ of negative streamers relative to the base oil, casting doubts about the validity of this hypothesis.

4.2.3. Electron Attachment Hypothesis

The electron attachment hypothesis of NPs is presented in [5]. It claims that NPs act as scavengers of electrons converting them into slower ions. The electron attachment process follows a charging relaxation time constant τ_r defined by:

$$\tau_r = \frac{2\epsilon_1 + \epsilon_2}{2\sigma_1 + \sigma_2'} \quad (3)$$

where ϵ is the electrical permittivity and σ is the electrical conductivity, with the subscript 1 for the host oil and 2 for the NPs. According to [5], this constant defines a criterion to estimate whether a NP can influence the streamer development. If τ_r is small compared to the time scale of streamer development, then the NP will relax fast enough and electrons will attach to it.

In the literature, this hypothesis has been claimed to disagree with the results presented in [3] where SiC nanofluids with a relaxation time constant of $\tau_r \approx 1$ ps yielded deleterious effects on the dielectric strength of the host oil. However, there are doubts whether the experimental sample preparation in [3] (including excessive moisture and size distribution of NPs) provides appropriate conditions to evaluate the effects of SiC NPs. On the other hand, the validity of the electron attachment

hypothesis has also been questioned based on results presented in [6,26]. They report that TiO₂ and SiO₂ nanofluids improve the dielectric strength of the host liquid despite of their relatively large τ_r . Nevertheless, it could be argued that water absorption of NPs is instead the main mechanism behind these reported improvements of SiO₂ and TiO₂ nanofluids, as already discussed in Section 4.2.1.

In order to revisit the validity of the electron attachment hypothesis only, it is here analyzed for negative streamer inception, which is not affected by moisture content. Interestingly, it is found that the changes in $V_{50\%}$ of negative streamers here reported for SiO₂, TiO₂, Al₂O₃ and ZnO are consistent with the relaxation time constant τ_r criterion. Thus, NPs with large τ_r do not have any appreciable effect on the $V_{50\%}$ of negative streamers, while only the ZnO nanofluids with low τ_r improve the $V_{50\%}$ of negative streamers even beyond the improvements perceived in the $V_{50\%}$ of positive streamers, as shown in Figure 10. This result is consistent with the electron attachment hypothesis judged by the charging relaxation time constant τ_r . Observe that the C₆₀ NPs cannot be used for this analysis due to the broad range of variation of its electrical conductivity. Using the electrical conductivities of C₆₀ NPs reported in the literature, ranging between 10⁻¹² S/m [37] and 1.7 × 10⁻⁶ S/m in [38] and [39], τ_r varies widely from less than one picosecond to several seconds.

5. Conclusions

Positive and negative streamer inception voltages are measured from ultra-sharp needles for SiO₂, TiO₂, Al₂O₃, surface modified ZnO and C₆₀ NPs dispersed in mineral oil at different mass fractions. It is found that NPs affect differently the inception of positive and negative streamers. While SiO₂, TiO₂ and Al₂O₃ NPs improves the 50% probability inception voltage ($V_{50\%}$) of positive streamers, they do not influence the $V_{50\%}$ of negative streamers. In turn, surface modified ZnO and C₆₀ NPs improve the $V_{50\%}$ under both polarities.

It is found that the hypothesis that NPs absorb water as postulated in [25,26] can explain the increments of the $V_{50\%}$ of positive streamers for the hydrophilic SiO₂, TiO₂ and Al₂O₃ NPs. Therefore, the here-reported experimental results support the hypothesis that hydrophilic NPs increase the positive streamer inception voltage of the base oil. Since hydrophobic surface modified ZnO and C₆₀ NPs also exhibit increments in the $V_{50\%}$ for positive streamers (relative to the host oil), it is concluded that other physical mechanism(s) should also be active.

On the other hand, the hypothesis that NPs increase the electron trapping and de-trapping processes in the liquid [6,27,40] fails to explain the presented results. Negative streamers in liquids are incepted through electronic avalanches in the liquid phase [10,11,36], and thus they should be influenced by the electronic properties of the liquid. Since results hereby reported show that TiO₂ NPs do not influence the negative streamers, it cannot be claimed that TiO₂ NPs influence the electron velocity of the mineral oil. On the other hand, the $V_{50\%}$ of negative streamers here reported for SiO₂, TiO₂, Al₂O₃, and ZnO nanofluids are consistent with the electron attachment hypothesis of NPs, judged through the relaxation time constant τ_r [5].

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