Improvement of transformer liquid insulation using nanodielectric fluids; a review

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Work aimed at enhancing transformer insulation performance by the use of nanodielectric fluids is reviewed, and future trends briefly discussed.

1. Introduction

During the last 20 years, the search for new applications for nanotechnology has become one of the busiest in science, engineering and manufacturing. New nanotechnology-based materials with superior properties have been developed and are already used in many everyday products and processes.

The application of nanotechnology to high-voltage (HV) engineering has been mainly oriented towards the development and characterization of the so-called nanodielectric materials [1]. In 1994, Lewis [2] suggested that "a major field of study in the future development of dielectrics will concern their properties when relatively few molecules are involved. Such smallness arises naturally at interfaces of nanometric thickness and will occur also when dielectrics are employed in the nano-technical devices of the future". The physical phenomena that govern the behaviour of materials at sub-microscopical scale are outlined in this publication [2]. The term "nanodielectric" was introduced by Frechette [3,4], who defined nanodielectrics as "multicomponent dielectrics possessing nanostructures, the presence of which results in the change of one or several of its dielectric properties".

During the last decade, the focus has been mainly on nanocomposites manufactured by adding nanoparticles (NP) to a solid polymer [1]. It has been widely reported that the dielectric properties of such materials are, in some aspects, superior to those of the matrix polymers, e.g., an increase in partial discharge inception voltage, suppression of space charge formation or enhancement of treeing life-time. The physical mechanisms involved on these changes are not yet fully understood, although they seem to be linked to the effect of the interfaces between the particles and the host polymers [4]-[9].

Recently, attempts have been made to apply nanotechnology to transformer insulating systems. A few authors have proposed the development of nanotechnology-based alternatives to traditional solid insulation [9,10]. However, most researchers seem to be working on the development of improved dielectric oils in which NP are incorporated in mineral oil (MO) or ester based liquids. These fluids are usually named nanodielectric fluids (NDF). They are two-phase systems composed of a solid dispersed NP phase and a liquid dispersion medium (base fluid) [11]. The term nanofluid was originally defined by researchers at Argonne National Laboratory (U.S.) as a "fluid in which nanometre-sized particles are suspended" [12]. They have been applied mainly to improve the heat transfer properties of the cooling liquids utilized in several industrial processes. It has been observed that the addition of NP to refrigerating fluids improves their thermal conductivities and their convective heat transfer performance [12].

Considering that the liquid insulation of transformers acts not only as an electrical insulator, but also as a refrigerant, it seems feasible that the application of nanotechnology could lead to liquids with superior properties which increase the reliability of the transformer. One of the first authors to suggest the application of nanofluids to transformers was Segal, who analysed the thermal and dielectric properties of a colloidal fluid containing mineral oil and magnetite nanoparticles [13,14]. As can be seen from the number of scientific works published in international journals and conferences (Figure 1), interest in NDF has risen rapidly over the last ten years. Some authors claim that the performance of NDF as insulating or cooling liquids is superior to that of the base refrigerant [15,16].
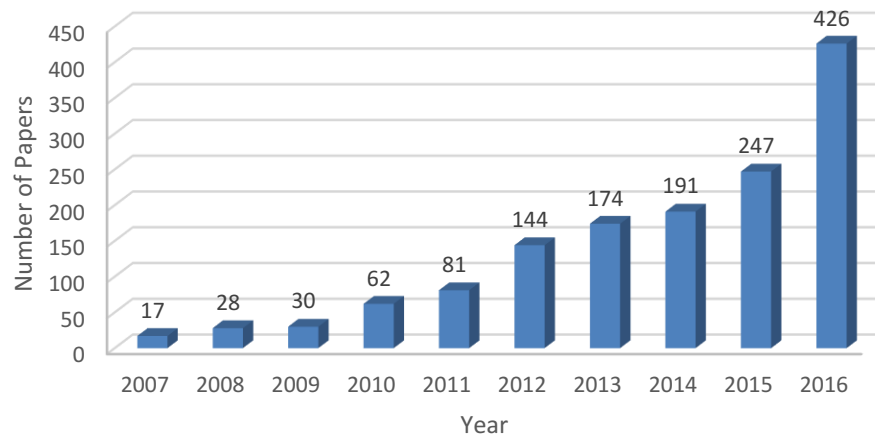

Figure 1.   Numbers of published papers on the subject "Nanofluids for application in transformers" from 2007 to 2016. (Based on Google Scholar).

It should be noted that NDF are not yet commercially available. Some problems remain to be solved, e.g., stability, interaction with the magnetic fields present in the transformer, and effect on the performance of the solid insulation within the transformer. Production costs must also be considered.

In this article we report on the state of the art on the electrotechnical use of NDF. The experimental results obtained by different authors, relating to the dielectric and thermal properties of NDF, are reviewed and compared. Practical problems such as the manufacture of the liquids, and basic theoretical aspects, are also discussed.

2. NDF SYNTHESIS
2.1 NDF Components

As stated above, NDF fluids are not yet commercially available. The results reported in the literature are mostly for fluids manufactured on a laboratory scale. Different types of NP and base fluid (in which the NP are dispersed) have been tested. Three main groups of NP have been used, namely (1) conductive, such as Fe_3O_4, or ZnO, (2) semi-conductive, such as TiO_2, Fe_2O_3, CuO or CuO_2, and (3) insulating, such as Al_2O_3, SiO_2 or SiO. Typical NP diameters are in the range 10-60 nm [17]-[20]. Most publications on NDF are concerned with MO-based fluids, but some work in which natural or synthetic ester-based oils were investigated has been published [21]-[27].

2.2 Preparation

The main difficulty involved in the preparation of NDF is the fact that, since NP are non-soluble compounds, stable colloids must be formed in order to suspend them in the base fluid. In the vast majority of cases the NP to be dispersed in the base fluid are prepared outside it, in a two-step manufacturing process. Such processes are popular because of their lower cost and because they do not require the use of specialized equipment. However, if the NP are directly added to the base fluids, sedimentation or agglomeration of the NP will occur, limiting the desired improvement in the properties of the NDF [24]. To prevent these unwanted effects, the NP are dispersed in the base fluid using ultrasonic agitation or magnetic stirring.

In most cases a surfactant is added to the base fluid/NP mixture, in order to reduce the repulsive forces between the NP and the base fluid, which in turn facilitates the formation of colloids within the base fluid and makes NP agglomeration less likely. Several surfactants have been used, e.g., hexadecyl trimethyl ammonium bromide for MO with magnetite (Fe_3O_4) NP [28], silane coupling agent Z6011 for MO with SiO_2 NP [11,29], and a non-ionic surfactant named Span80 composed of fatty acids for MO with TiO_2 NP. The most widely used surfactant is oleic acid [30]-[33].

After the mixing process, the NDF is dried and degassed in order to eliminate bubbles and moisture. Mixing usually involves heating under reduced pressure, typically at 60ºC and 1kPa for 24-72 hours [15, 34 ,35]. Degassing NDF can be more difficult than degassing conventional MO. In order to remove impurities, the NDF may be passed through a membrane filter which captures particles larger than its pore size [36].

In the one-step method the NP are simultaneously synthesized by physical vapour deposition (PVD) or a liquid chemical method, and dispersed within the base fluid. This method results in less NP agglomeration [37], but is not readily implemented except in a specialized laboratory.

2.3 Stability

One of the main problems with NDF is to ensure long-term dispersal of the NP within the base fluid. Dispersal times varying from hours to several months have been reported [24, 30, 31, 38, 39]. The dispersal times depend on the NP concentration, e.g., for MO with SiO_2 NP, NP concentrations of 0.01, 0.02 and 0.1 vol. % lead to dispersal times of 1 month, 2 days and less than 24 hours respectively [39]. Atiya et al [40] showed that, for a MO with TiO_2-based NF, concentrations of the surfactant cetyltrimethylammonium bromide, surfactant above 0.3 vol. % increased the NP agglomeration. It has also been reported that the addition of large amounts of surfactant can reduce the dispersal time and degrade the dielectric properties of NDF [41].

2.4 Safety

Several agencies have expressed concern about the potential health hazards which nanomaterials can pose for humans [41]. However, given the available toxicological data, the European Commission has suggested that many of them are not dangerous in moderate doses [42]. NP can enter the human body through inhalation, reaching the lungs, other organs and the central nervous system [43]. NP can also enter the body through the digestive system, due to poor hygiene practices, or through the skin. The risks are particularly severe when working with NP powder, which is the most widely form of NP used in NDF manufacturing processes. At present it is recommended that the procedures commonly adopted when working with ultrafine particles in the laboratory be followed [43], e.g., use of masks, safety glasses, gloves, protective clothing and security chambers.

3. Dielectric Properties

Most of the work published on NDF is concerned with their dielectric properties, particularly in comparison with the corresponding properties of the base fluid. Most authors report that dispersion of low NP concentrations in a base fluid improves the performance of the base fluid. However, the reasons for the improvement are not always clear.

Several authors have suggested that, when an NDF is subjected to an electric field, positive ionic charges accumulate on the surface of the NP, attracting negative ionic charges around them [29,36]. Thus, an electrical double layer (EDL) is formed, as shown in Figure 2. The volume of the EDL close to the NP surfaces is called the compact layer, and consists of immobile negative ions strongly attracted 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 [34]. The formation of the EDL in a NDF subjected to an electric field is believed to be the main reason for the improvement in dielectric properties reported by many authors [34, 44].

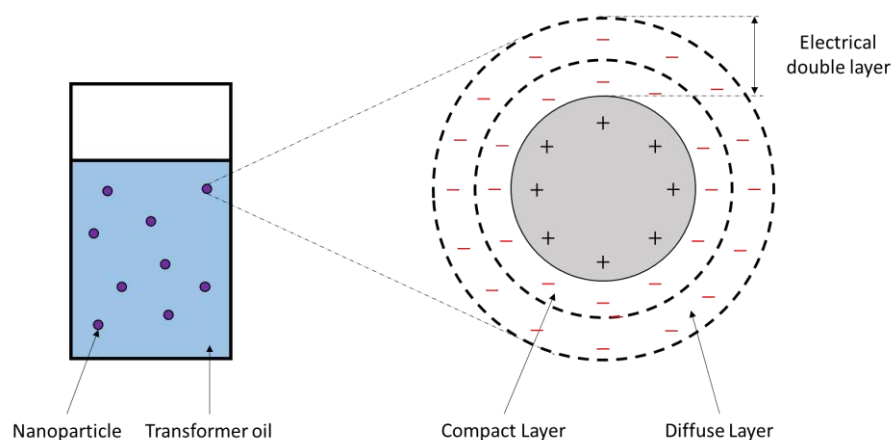*Figure 2.   Electrical double layer formation in NDF. Adapted from* [29],[34].

In order to quantify the improvement in the dielectric properties of the base fluid due to the addition of NP, Hwang [45] defined the charge relaxation time τ of a NDF, which is a measure of the extent to which the times and frequencies of electrodynamic processes in the base fluid are affected by the presence of the

NP. τ depends on the DC conductivity (σ) and low frequency permittivity (ε) of the NP and the base fluid, and is given by

$$\tau = \frac{2\varepsilon_{fluid} + \varepsilon_{NP}}{2\sigma_{fluid} + \sigma_{NP}}$$

If the relaxation time of the NP is shorter that the time scale of the electrodynamic process being considered, e.g., dielectric breakdown, the effect of the NP on the process will be very noticeable. On the other hand, if the relaxation time is longer than the time scale of the process the NP will have little effect on the breakdown processes. Calculated relaxation times for four types of NP dispersed in mineral oil are shown in Table 1 [47]; they differ greatly. Assuming the time scale of dielectric breakdown in MO to be typically 10^{-9} s we would then expect Fe_3O_4 and ZnO to affect the breakdown greatly, but Al_2O_3 and SiO_2 to have little effect. This is inconsistent with some published experimental data, e.g., increases of as much as 30% in the AC-BV of MO following the addition of Al_2O_3 and SiO_2 NP (as shown in Table 2).

Table 1: Charge relaxation times for four types of NP dispersed in mineral oil (MO).
$\sigma_{MO}=1\cdot10^{-12}$ S/m [45], $\varepsilon_{MO} = 2.2\cdot\varepsilon_0$ [45], $\varepsilon_0=8.854 \times 10^{-12}$ F/m. [47]

	Magnetite Fe_3O_4	Zinc oxide ZnO	Alumina Al_2O_3	Silica SiO_2
Electrical conductivity [S/m]	10^4	10	10^{-12}	1.4×10^{-9}
Relative permittivity	80	7.4	9.9	3.8
Charge relaxation time [s]	7.47×10^{-14}	1.05×10^{-11}	42.2	5.18×10^{-2}

3.1 AC Breakdown Voltage

AC breakdown voltage (BV) has been measured by many authors. Most papers present measurements performed in accordance with IEC 60156 standard [46].

3.1.1 Effect of NP conductivity on AC-BV

In Table 2, the results reported by various authors for various NDF, obtained by dispersing conductive, semi-conductive and insulating NP in MO or natural ester (NE) oil, are compared. (Some authors did not specify the NP concentrations). The average AC-BV for dry mineral oil is 70.3 ± 16.7 kV [47]. Some increase in AC-BV was reported in all cases. A possible explanation is that electrons are trapped by the NP, which slows up the propagation of the breakdown streamer and thus increases the breakdown voltage [19].

Table 2. Increase in AC-BV of NDF containing various NP in mineral oil or natural ester (NE) oil.

NP/ Base fluid	Conductivity	Average NP size (nm)	NP loading	% Increase in AC-BV	Ref.
Fe_3O_4/MO	Conductive	10	-	42.8 %	[49]
Fe_3O_4/NE	Conductive	30	-	19.8 %	[50]
Fe_3O_4/MO	Conductive	10	-	44.3 %	[17]
ZnO/MO	Conductive	34	0.0005 vol. %	8.3 %	[49]

TiO₂/NE	Semi-conductive	20	0.00625 vol. %	31 %	[22]
TiO₂/MO	Semi-conductive	<20	0.00075 vol. %	15 %	[51]
TiO₂/MO	Semi-conductive	10	-	33 %	[17]
SiO₂/MO	Insulating	15	0.01 vol. % 0.02 vol. %	19 % 26 %	[52]
SiO₂/MO	Insulating	15	0.0074 vol. %	17 %	[48]
Al₂O₃/MO	Insulating	10	-	35.5 %	[17]

3.1.2 Effect of NP concentration on AC-BV

One might expect that the AC-BV would increase with increasing NP concentration. However, this is true only up to certain concentration limits, beyond which the AC-BV decreases [15]. Table 3 shows the results of a study [51] in which increasing concentrations of TiO_2 NP were dispersed in MO. Disregarding the "rogue" result at 0.003 g/L, the AC-BV increased with increasing NP concentration up to 0.006 g/L and decreased at higher concentrations, becoming less than the AC-BV of MO at 0.05 g/L. The same effect has been reported in [53] and [54], but with different optimal NP concentrations. It was suggested in [55], that the reduction in AC-BV at higher NP concentrations might be due to the accumulation of conductive NP chains near the electrodes, facilitating discharges.

Table 3. AC-BV of a NDF consisting of various concentrations of semi-conductive TiO₂ NP (diameter <20 nm) dispersed in MO [51].

Samples	Mean AC-BV (kV)	% Increase in AC-BV
Mineral Oil	71.59	0
TiO₂ (0.003g/L)	67.33	-5.9
TiO₂ (0.006g/L)	82.48	15.2
TiO₂ (0.01g/L)	77.82	8.7
TiO₂ (0.03g/L)	71.76	0.2
TiO₂ (0.05g/L)	53.25	-25.6

The evolution of the AC-BV of NDF with NP concentration is similar for conductive, semi-conductive and insulating nanoparticles, and the optimal NP concentrations seem to be dependent on the particle type, the nature of the BF and the manufacturing process [51]-[55].

3.1.3 Effect of moisture in the base fluid on AC-BV

It is well known that the presence of moisture degrades the dielectric properties of oil. Several authors have suggested that such degradation might be eliminated, or at least significantly reduced, by dispersing NP in

the oil [11],[12],[15]. Jin [11] measured the AC-BV of MO and a MO-based SiO_2 NDF with moisture levels of 15 and 25 ppm. The SiO_2 NP concentrations were 0.01 and 0.02 vol. %, and the NP sizes were in the range 20-30 nm. As shown in Table 4, the AC-BV increased with increasing NP concentration at a given water concentration, but decreased with increasing water concentration at a given NP concentration. He suggested two phenomena which might lead to these results. These are:

(1) water molecules seem to be attracted to charged conductive NP, and cluster around them. The mobility of the water molecules would therefore decrease, but although the presence of water reduces the AC-BV, the reduction would be smaller in the presence of the NP

(2) some semi-conductive or insulating NP are hydrophilic so that, although they are not conductive, they would bind on their surfaces some of the water molecules dispersed in the oil.

Jin [11] compared the AC-BV of two MO-based NDF, one containing hydrophilic silica NP and the other containing silica NP modified with the surfactant silane coupling agent Z6011, which renders them hydrophobic. The AC-BV of the latter NDF was similar to that of the base fluid, whereas the AC-BV of the NDF containing hydrophilic silica NP was 10-25 % greater than that of the base fluid (as shown in Table 4), presumably because water molecules were bound to the surfaces of the hydrophilic NP.

Table 4. AC-BV of MO with two moisture concentrations and two insulating hydrophilic SiO_2 NP concentrations. The NP sizes were in the range 20-30 nm [11].

Samples	Mean AC-BV (kV)	% Increase in AC-BV relative to MO with the same moisture content
MO, 25 ppm moisture	55	-
MO, 15 ppm moisture	80	-
MO, 0.01 vol. % SiO_2, 25 ppm moisture	66	20
MO, 0.01 vol. % SiO_2, 15 ppm moisture	88	10
MO, 0.02 vol. % SiO_2, 25 ppm moisture	69	25
MO, 0.02 vol. % SiO_2, 15 ppm moisture	91	14

Weijun Yin [15] reported that, in a MO-based NDF containing Al_2O_3 NP, the AC-BV decreased with increasing water content, but a much greater water content was required in samples containing NP to produce a comparable decrease in AC-BV, relative to samples without NP (Figure 3).
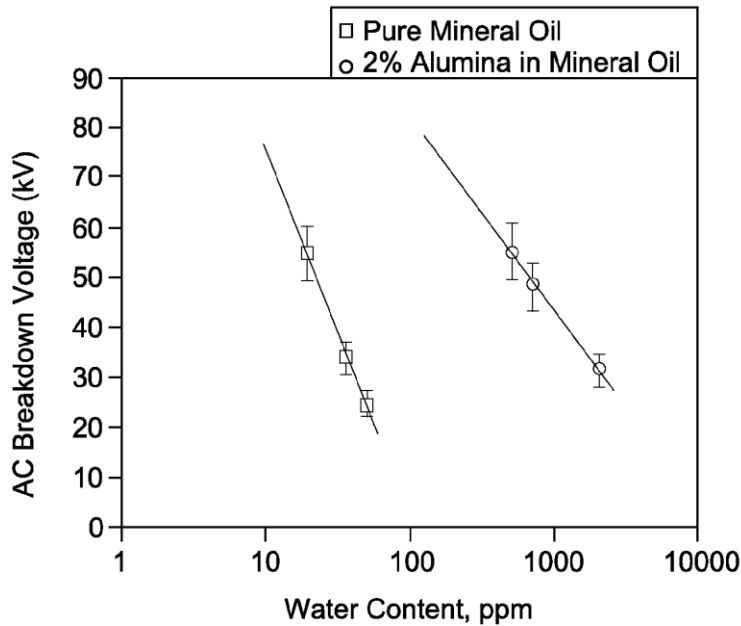
Figure 3. Effect of water on the AC-BV of MO and an MO-based NDF containing 2 vol. % of Al$_2$O$_3$ NP [15].

3.1.4 Effect of NP size *on AC-BV*

In [32], several natural ester-based NDF were studied. The natural ester was FR3, and the NDF were manufactured by dispersing Fe$_3$O$_4$ NP of sizes 12 nm, 21 nm and 41 nm, to a concentration of 0.03 vol. % in the FR3, and using oleic acid as surfactant. A range of AC-BV was obtained; its mean value increased from 55.1 kV for the FR3 alone to 68.5 kV for the NDF containing the 41 nm NP (Figure 4). The authors suggested that the increase of the AC-BV with increasing NP size might be due to the fact that the ability of Fe$_3$O$_4$ NP to trap electrical charge carriers increases with increasing NP surface area.

According to Liu et al [55], space charge density in NDF decreases quickly following application of electrical stress, and thus the internal electric fields appearing within them are lower than those that would appear in the base fluid under the same stress. However, the addition of larger NP would cause field distortion that might degrade the dielectric properties of the fluids, i.e., a decrease in AC-BV at larger NP sizes might be expected.
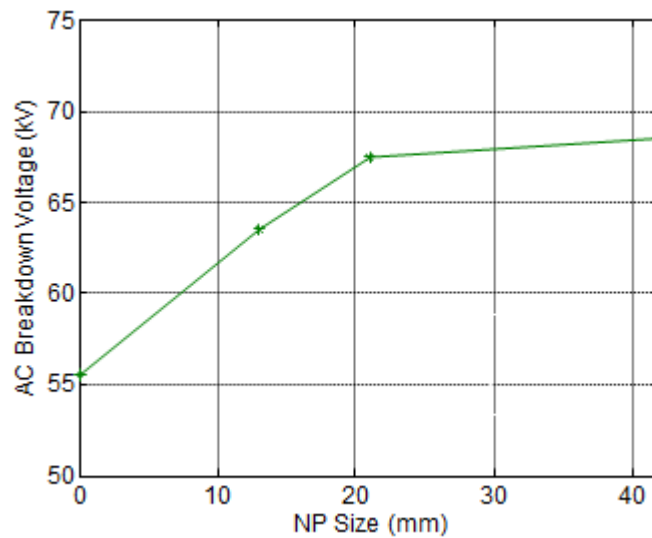
Figure 4.   Influence of Fe$_3$O$_4$ NP size on the mean AC-BV of FR3-based NDF [32].

3.1.5 Effect of thermal ageing on AC-BV

The effects of thermal ageing of NDF on their AC-BV have been studied [56]-[59]. Karthik et al. [56] synthesized three types of NDF, all based on MO. The dispersed NP were Fe$_3$O$_4$ (magnetite) with particle size 10-50 nm, SnO (10-50 nm) and insulating SiO$_2$ (5-20 nm) at a concentration of 0.2 g/l. Oleic acid was added as a surfactant to the Fe$_3$O$_4$ NDF, but not to the second and third NDFs. The NDF were poured into hermetically sealed vessels (with an air head) and aged for 675 hours (approximately one month) at a temperature of 120°C. Figure 5 shows the average value of the AC-BV (extracted from the Weibull distribution fitting) for the MO and for each of the three NDF. Addition of each of the three types of NP increased the AC-BV of the MO. The increases persisted after the thermal ageing. The AC-BV reductions after aging in the MO and in the NDF containing Fe$_3$O$_4$ were approximately 60 % and 50 % respectively. The corresponding reduction in the NDF containing SnO was about 10 %, and zero within experimental uncertainty in the NDF containing SiO$_2$. The large reduction in the NDF containing Fe$_3$O$_4$ was not explained in detail, although the authors [56] noted that only the NDF was surfactant-treated. Similar results were reported in [57] for a MO-based NDF containing TiO$_2$ NP subjected to thermal ageing.
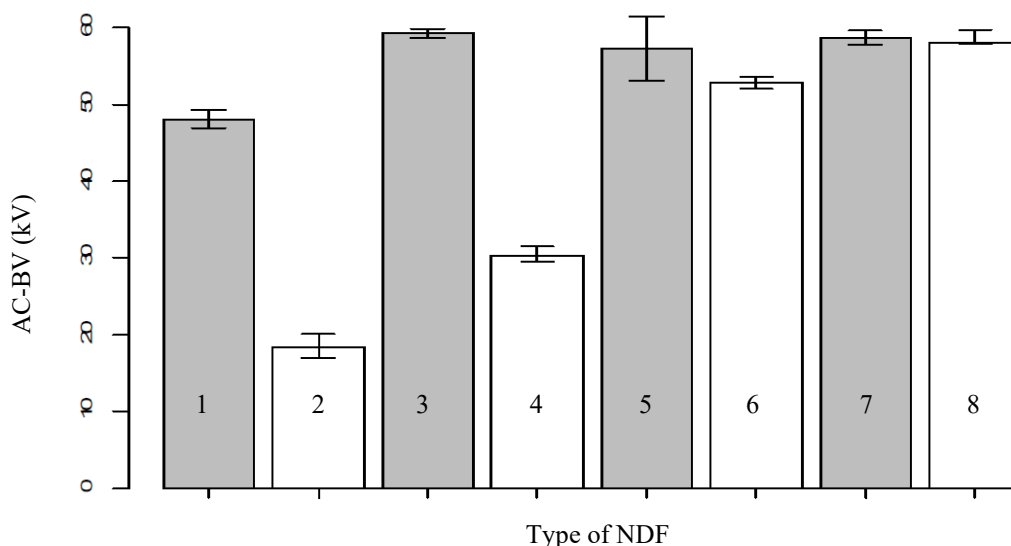
Figure 5. AC-BV a*verage values for three MO-based NDF before and after thermal aging at 120 ^0C for 675 hours in air* [56]. *1-MO; 2-Aged MO; 3-Magnetite NDF; 4-Aged Magnetite NDF; 5-SnO NDF; 6-Aged SnO NDF; 7-SiO$_2$ NDF; 8-Aged SiO$_2$ NDF. The experimental uncertainties in the AC-BV values are indicated.*

Zhi-Feng et al [57] suggest that the improved AC-BV of aged MO-based NDF compared to aged MO is due to the fact that a fraction of the molecules generated as by-products of the ageing reactions can bind to the NP surfaces. Consequently the internal electric field distribution within the aged NDF is more uniform than the corresponding distribution in MO aged in the same manner.

3.2 DC Breakdown Voltage

Kudelcik et al. [60] reported an increase of the positive DC-BV of MO-based NDF containing Fe_3O_4 NP with diameter 10 nm. The DC-BV increased with increasing NP concentration up to 1 vol. %, but decreased sharply when the concentration was increased to 2 vol. % (Figure 6). This variation with NP concentration resembles that of the AC-BV presented in section 3.1.2. The authors give two possible reasons for the initial increase in DC-BV with increasing NP concentration. Firstly the NP act as electron traps, limiting the concentration of "fast" electrons in the oil and generating "slow" negatively charged NP. The latter slow down the ionization processes which precede breakdown. Secondly the presence of NP can affect the process of bubble formation within the NDF, since the permittivity of the NP is higher than that of the surrounding liquid. Under an applied voltage the charged NP move towards the areas of maximum electrical stress, where most of the bubbles are generated. The authors also suggest that, at higher NP concentrations, local electric field gradients can lead to lower BV.
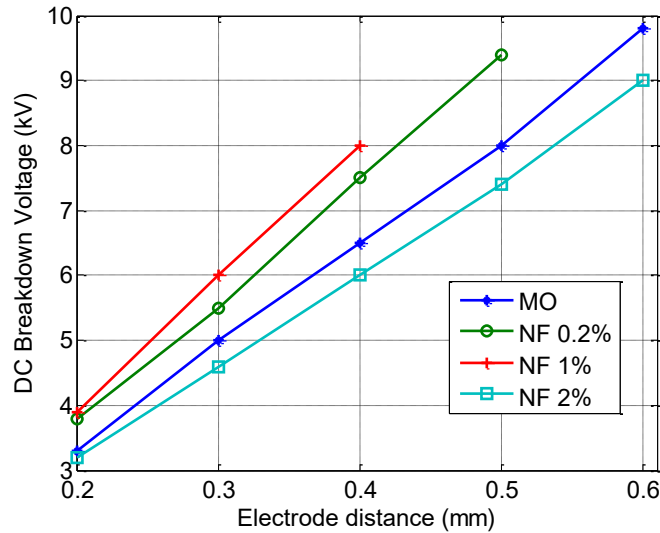
Figure 6.   DC-BV of MO and MO-based NDF containing three different concentrations of Fe₃O₄ NP, as functions of the electrode separation [60].

The same authors [60] subjected the NDF to an external magnetic field of 20 mT, which reduced the DC-BV. They suggested that the reason could be the formation of micrometre-sized Fe_3O_4 NP agglomerations in the NDF as a result of magnetic dipole-dipole interactions between NP.

Du [35] investigated the effect of polarity on the DC-BV (one grounded electrode) of a MO-based NDF containing 0.075 vol. % of TiO_2 NP. Relative to the unmodified MO, the DC-BV fell by 8 % under positive polarity, but increased by 28 % under negative polarity. The authors suggest that electron emission is more "violent" under negative polarity than under positive polarity, but do not explain the meaning of "violent" in this context.

4. Streamer propagation in NDF

A streamer is a transient electrical discharge which may appear when an insulating medium, such as insulating oil, is exposed to a large voltage drop, such as a lightning or switching impulse. Streamer propagation is generally measured using a standard lighting impulse voltage 1.2/50 μs in a needle-sphere combination of electrodes with a 25 mm gap. The speed of streamer propagation in dielectric fluids is strongly dependent on the polarity of the applied voltage and its peak amplitude [62]. In the particular case of MO it has been observed that streamers originating from a positive electrode tend to initiate at lower voltages than those originating from a negative electrode, and to propagate faster and further [62].

Several authors have studied streamer propagation in a number of NDF, comparing their performance with that of the base fluid [17], [19], [21], [38], [50], [63], [64]. They concluded that, under positive polarity (positive streamers), NDF tend to have lower streamer velocities and higher streamer BV than the base fluids, when high densities of high energy electrons are generated. On the other hand, under negative polarity the ability of NDF to withstand over-voltages does not differ appreciably from that of the base fluid.

The formation mechanisms of streamers in NDF have been studied using finite elements analysis (FEA) [19], [45], [61], [65]. It is believed that the low mobility of the NP hinders the development of a space charge zone at the streamer tip, thereby suppressing the propagation of the electric field wave that is needed to drive electric-field-dependent molecular ionization, and hence streamer propagation [65]. MO-based NDF exhibit lower space charge densities than pure MO, due perhaps to faster negative charge decay rates in NDF [65].

The trapping of fast electrons by slow NP seems to be the cause of the decrease in positive streamer velocity mentioned above.

A broad theoretical study of streamer propagation in NDF has been published by O'Sullivan [19]. Several models were are described and implemented using FEA. The physical phenomena that take place when a NDF is subjected to an impulse voltage were analysed. The author concluded that, since streamer propagation in insulating fluids requires ionization of the fluids and, since many of the electrons generated by this ionization are trapped by the NP before they are transported out of the ionization region, the time scales of streamer development in NDF will be significantly longer than those in the unmodified base fluids.

The effect of the NP on the propagation velocity of positive streamers in an MO-based NDF containing Fe_3O_4 NP has been studied in [15]. The velocities decreased by as much as 46 % compared to MO. A slower propagation velocity implies a greater probability that the impulse voltage will be extinguished before the oil breaks down, so the positive streamer BV will be higher than in unmodified MO. Hwang [63] showed that the higher the NP conductivity, the smaller the reduction in the positive streamer propagation velocity.

The average BV for positive lightning impulses in a natural ester (NE) was found to be 73.9kV, while for an NE-based NDF containing Fe_3O_4 NP (1 vol. %) it was 101.5 kV [23]. The corresponding BV for negative lightning impulses were 83.8 kV and 93.7 kV respectively. It has also been reported [50] that the breakdown time measured in a similar NDF (based on a similar NE with Fe_3O_4 NP) was longer for the NDF than for the base fluid (12.0 µs and 9.9 µs respectively).

Lv et al. [66] investigated an MO-based NDF in which TiO_2 NP (10 nm) had been dispersed (0.075 vol. %). In Table 5 the positive and negative impulse breakdown voltages are presented. Positive streamers pose a greater risk for MO than negative streamers, since under positive streamers dielectric breakdown occurs at lower voltages values and at shorter breakdown times, e.g., at 97.16 kV for positive streamers and at -108.46 kV for negative streamers.

Table 5. Positive and negative impulse breakdown voltages and times-to-breakdown for unmodified MO and an MO-based NDF in which 10 nm TiO_2 NP had been dispersed (0.075 vol. %) [66].

Positive impulse breakdown				
Samples	BV (kV)	Standard deviation (kV)	Time-to-breakdown (µs)	Standard deviation (µs)
Unmodified MO	74.27	5.34	13.18	0.61
NDF	97.16	6.05	25.65	1.70
Negative impulse breakdown				
Unmodified MO	-116.42	3.92	49.58	6.43
NDF	-108.46	5.23	11.39	3.77

5. Partial discharge in NDF

Breakdown occurs after discharge initiation and propagation through the oil. The BV measurement alone provides little information on the discharge process. It is of interest to investigate the differences between the discharge mechanisms in the base fluid and the associated NDF, and one approach is to measure the Partial Discharge (PD) activity in both.

The PD inception voltage (PDIV), time duration, rise-time, total discharge magnitude and PD voltage amplitude for MO, and MO-based NDF containing silica and fullerene NP (average particle size 15 nm and 1 nm respectively), under DC voltage, are compared in [67]. The discharge mechanism in MO depends strongly on the polarity of the applied DC voltage. Under negative polarity the PDIV and the total discharge magnitude were similar for MO and the NDF, but under positive polarity the PDIV increased and the total discharge magnitude decreased relative to the MO values. The authors suggested that the improved dielectric performance of the NDF could be due to the trapping of electrons, water molecules, acids, or other additives on the surface of the NP, and the subsequent slowing down of the breakdown processes [67].

It was reported [28],[35] that the PDIV of MO increased when TiO_2 NP were added, and the PD magnitudes and pulse rates decreased (Table 6). It was also reported that, as expected, increasing the applied voltage from PDIV to 2 x PDIV increased the discharge magnitude and the number of pulses in the MO and the NDF. Figure 7 shows phase-resolved-partial-discharge (PRPD) patterns measured in a 25 mm needle-sphere gap, the needle tip having 50 μm radius of curvature, for applied voltages of (a) PDIV and (b) 2 x PDIV [35].

Table 6. PDIV, PD magnitudes, and numbers of PD pulses measured in MO and in MO - TiO_2 NDF at 1.0 and 2.0 PDIV, during 10 minute measurement intervals [28].

Sample	PDIV (kV)	Std. deviation (kV)	Measured at PDIV		Measured at 2 x PDIV	
			Pulses /10 s	Mean discharge magnitude (pC)	Pulses /10 s	Mean discharge magnitude (pC)
MO	30.66	2.66	9	435	245	6062
MO-TiO_2 NDF	33.13	1.79	6	245	168	5180

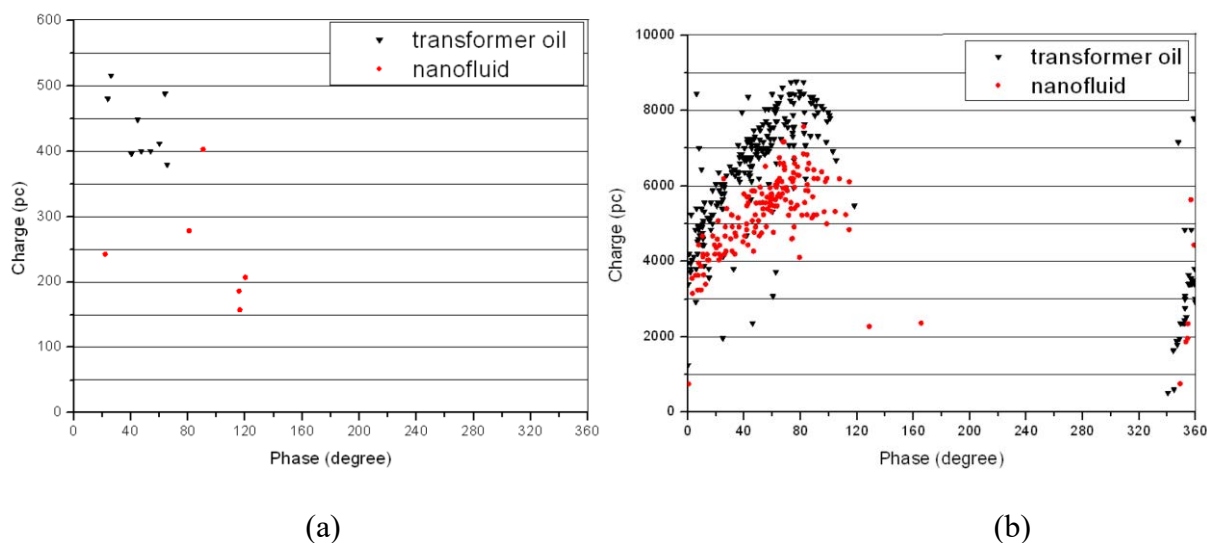(a)   (b)

Figure 7. Phase-resolved-partial-discharge (PRPD) patterns measured in MO and in MO - TiO_2 NDF in a 25 mm needle-sphere gap, the needle tip having 50 μm radius of curvature, for applied voltages of (a) PDIV and (b) 2 x PDIV [35].

Three MO-based NDF, containing silica ferrofluid, graphene oxide, and SiO$_2$ NP were tested under DC and AC voltages and their performances compared with that of MO [68]. The NP concentrations were in the range 0.1 - 0.2 g/l. Under AC voltages all three showed an increase in PDIV compared to MO, but under both positive and negative DC voltages the measured PDIV were lower than those observed under AC (Figure 8).
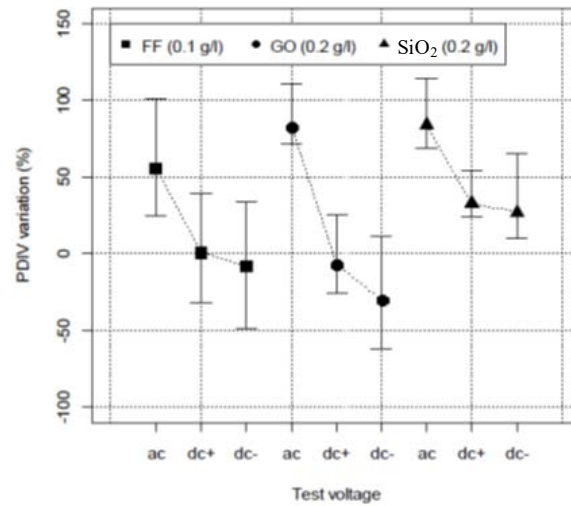

Figure 8. *PDIV for three MO-based NDF under AC and positive and negative DC voltages. The nanoparticles were FF = silica ferrofluid, GO = graphene oxide, and SiO$_2$ [68].*

Liu et al [55] observed an increase of 20% in the PDIV of an MO-based NDF containing 0.12 vol. % concentration of aluminium nitride NP, and suggested that the increase could be due to the trapping of electrons on the NP surfaces. They stated that partial discharges are always caused by a localized increased electric field strength in the volume or on the surface of a defect, e.g., water, bubbles or particles, the increased field being due to differences in relative permittivity between the defect and the host material. Although the presence of charged particles in transformer oil generates local electric field distortion within the oil which can lead to the formation of partial discharges, this is not what happens when NP are dispersed in oil [22], [55]. According to the authors of these publications, when NP are dispersed in an insulating fluid the charge density within the NDF will decrease rapidly with increasing time under an externally applied voltage, and the internal electric field will therefore be lower than that in the base fluid under the same applied voltage. In [22] the authors suggested that the rapid decrease in space charge density could be attributed to a high shallow trap density created by the NP, and hopping transport of charge between those traps. Rapid transfer of charge carriers through the shallow traps implies increased mobility of the charges in the NDF, compared to the base fluid, which in turn reduces the accumulation of space charge [69] and distortion of the electric field distribution. The end result is an increased partial discharge inception voltage and a decreased partial discharge probability.

6. Thermal Properties

Transformer insulating oil also acts as a cooling agent. In that context, its thermal conductivity is important, as are its viscosity and density. As mentioned in the Introduction, the addition of NP was initially regarded as a way of improving the thermal properties of cooling liquids used in different industrial processes [12]. Consequently, many publications in this field deal with improvement in the thermal properties of liquids such

as water, ethylene glycol and engine oil when NP are dispersed in them. We consider these first, describing them as nanofluids rather than NDF.

6.1 Factors influencing the thermal conductivity of non-MO-based nanofluids.

The results obtained by several groups of authors on the thermal properties of various nanofluids were reviewed in an extensive report [12] published in 2007. These authors compared the thermal properties of nanofluids fabricated using different base fluids, e.g., water, ethylene glycol and engine oil, and several types of NP. Improvement in the thermal properties was observed in most of the nanofluids, relative to the base fluids, but the magnitude of the improvement was dependent on several factors [70]-[77].

We consider now the influence of several factors on the thermal conductivity of various nanofluids which have been proposed as coolants with superior performance in a range of applications. Usually the thermal conductivity of a nanofluid is more sensitive to temperature than its base fluid [71], and in consequence the reported improvements are more noticeable at higher temperatures.

6.1.1 Influence of the choice of base fluid
Xie et al. [74] compared the increases in the thermal conductivity obtained by dispersing aluminium oxide NP in three different base fluids, namely water, ethylene glycol and pump oil. The highest percentage increase was observed in the base fluid with the lowest thermal conductivity, i.e., pump oil, whereas the lowest percentage increase was observed in water, which has the highest thermal conductivity of the three base fluids.

Ozerinc et al [76] state that, according to the "renovated" Maxwell model [78], the thermal conductivity ratio of a nanofluid (thermal conductivity of the nanofluid divided by the thermal conductivity of the base fluid) increases as the thermal conductivity of the base fluid decreases. However, they emphasize that the effect of the NP on other physical properties of the base fluid, e.g., its viscosity, should be considered simultaneously with their effect on the thermal conductivity.

6.1.2 Influence of NP type and concentration

Measurements of the thermal conductivity of nanofluids obtained by dispersing metallic or oxide NP in various cooling agents, mainly water, ethylene alcohol and engine oil, indicated that:

(a) Metallic NP, such as Cu or Fe, increase the thermal conductivity of the base fluid, even at very low concentrations. Concentrations below 0.5 vol. % may cause increases in the range 10-15 %, and concentrations of 2.5 vol. % more than 115% [72]. However, metallic NP are not readily available, and tend to oxidize during manufacture or manipulation [12].

(b) Low thermal-conductivity NP, e.g., Al_2O_3, CuO, TiO_2, cause very small thermal conductivity increases at concentrations below 2 vol. %. At concentrations in the range 4-10 vol. %, increases conductivity around 30% were observed in water-based nanofluids with Al_2O_3 and CuO NP, and in ethylene glycol-based nanofluids with CuO NP [72]-[74]. (At these concentrations the increase in the viscosity of the NF relative to the base fluid in all cases was less than 30 % [12]). Concentrations of CuO NP up to 10-15 vol. % in ethylene-glycol yielded increases of 40-50 % in the thermal conductivity relative to the base fluid [72]-[75]. Nanofluids containing NP concentrations greater than 15 vol. % tend to be unstable, because the NP tend to agglomerate and precipitate. Some authors expect a reduction in thermal conductivity at such NP concentrations [12].

For oxide-type NP, the thermal conductivity of the NP seems to have very little influence on the increase in the thermal conductivity of the nanofluids compared with the base fluids (water, ethylene glycol and pump oil) [12].

6.1.3 Influence of NP size

The increase in thermal conductivity achieved by the dispersion of NP with diameters in the range 30-60 nm in several base fluids, e.g., ethylene glycol, pump oil and water, has been studied by several research groups [72]-[74]. There seems to be general agreement that the bigger the NP the bigger the increase. However, the effect of NP agglomeration, which tends to occur more readily with larger NP, was not considered by the authors of these publications.

6.1.4 Influence of NP shape

It seems that cylindrical NP cause larger increases in the thermal conductivity of the base fluid than spherical NP. Xie et al [74] and Murshed et al [75] found that the addition of cylindrical NP (SiC and TiO_2) to water and ethylene glycol resulted in an increase in thermal conductivity which was 5-10 % greater than the corresponding increase following the addition of spherical SiC and TiO_2 NP at similar concentrations to the same base fluids.

6.2 Thermal properties of mineral oil- and ester-based NDF

Few publications deal with the thermal properties of transformer-oil-based NDF containing NP [24],[25], [39], [70], [79], [80]; more importance is placed on their dielectric properties. Two publications have been concerned with the temperature distributions within a transformer when MO-based and ester-based NDF were used as insulation [81],[82]. We consider now the main conclusions presented in the first group of publications.

6.2.1 Thermal conductivity

Nadolny et al [24] report on an experimental study in which the thermal conductivities of different MO-based NDF (TiO_2 and fullerene-C60 NP) were measured and compared with that of MO. Increases smaller than 1 % were obtained for each of the NDF. Similar results were obtained by Jin et al. [78] for MO-based NDF with various concentrations of SiO_2 NP ((a) (a)), and by Mansour and Elsaeed [82] for an MO-based NDF with 0.1 vol. % of Al_2O_3 NP. These small increases in thermal conductivity are consistent with the small increases in non-MO based nanofluids containing low NP concentrations (Section 6.1).

Du et al [79], [83] carried out an experimental study in which some of the dielectric and thermal properties of two MO-based NDF containing boron nitride (BN) and Fe_3O_4 NP were analysed. Although the increase in the thermal conductivity was below 2% (relative to MO) in all cases ((a) (b)), the NDF showed higher thermal conductivity than the MO. The dielectric properties of both types of NDF were also superior to those of the MO.

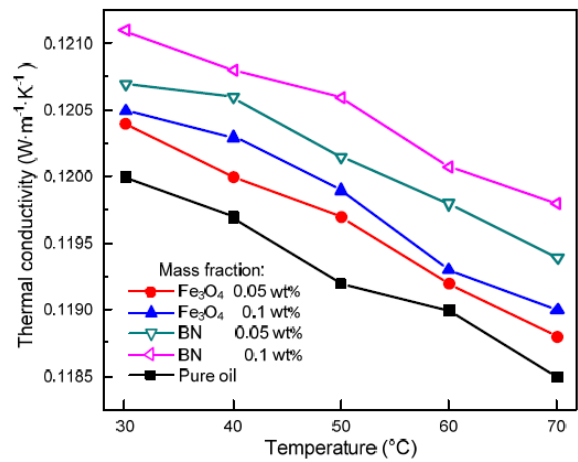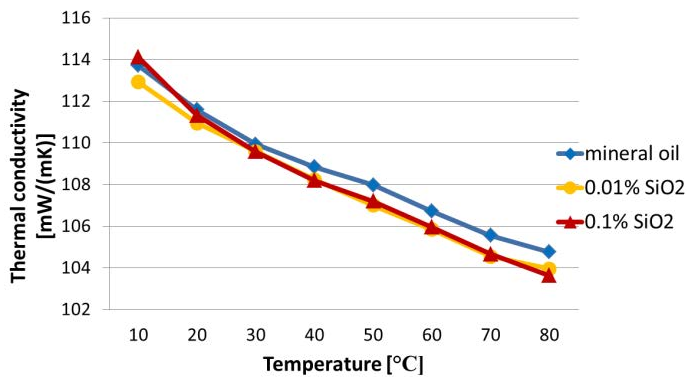(a)   (b)

Figure 9. Temperature dependence of the thermal conductivity of MO-based NDF containing (a) SiO₂ nanoparticles [78] (concentrations in vol %), (b) boron nitride (BN) and Fe₃O₄ nanoparticles [83] (concentrations in weight %).
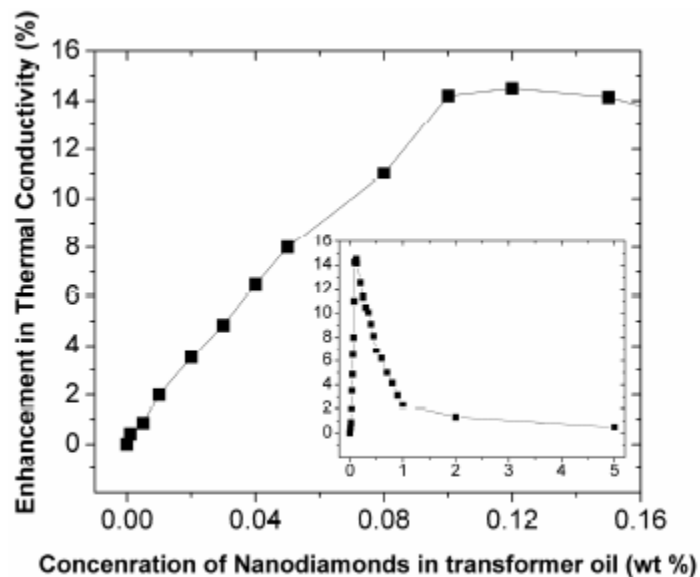

Figure 10. Increase in thermal conductivity of an MO-based NDF containing various concentrations of nano-diamond nanoparticles (concentrations up to 0.16 % wt in the main graph and up to 5 % in the rectangular inset) [39].

We have found only one publication [39] reporting a significant increase (15 %) in the thermal conductivity of an NDF (nano-diamond NP at a concentration 0.1 vol. % in MO) compared to that of the MO (Figure 10). It may be that the hot wire method used in [39] to measure the thermal conductivities yielded unrealistically high values [70].

6.2.2 Viscosity and density

Viscosity and density can influence the thermal performance of cooling liquids. Several authors have studied the effect of NP on the viscosity and density of the base fluids, finding that the effect of NP on the density and viscosity of MO and esters, at typical NP concentrations below 1 % vol, was very small [24],[25],[39],[79]. Nadolny et al [24] made viscosity and density measurements on an MO-based and a synthetic ester-based NDF containing fullerene C60 NP, and TiO_2 NP coated with the surfactant Span 20. Figure 11 shows the viscosity of some of the NDF based on MO, at temperatures between 25 and 80 ºC. In order to study the influence of the surfactant Span 20 the authors [24] also made measurements on mixtures of MO and surfactant at a concentration 5g/l. They found that the viscosity increases were in all cases less than 8 %.
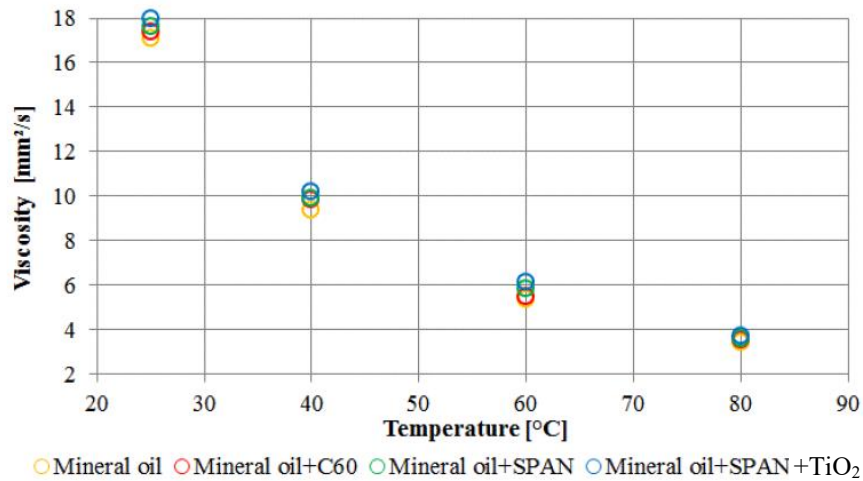

○ Mineral oil ○ Mineral oil+C60 ○ Mineral oil+SPAN ○ Mineral oil+SPAN+TiO₂

Figure 11. Viscosity vs. temperature measured on two MO-based NDF containing C60 NP or TiO_2 NP, on a mixture of MO and the surfactant Span, and on the MO [24].

Table 7. Viscosity and density vs. temperature measured on two MO-based NDF, one containing C60 NP and the other TiO_2 NP and the surfactant Span, on a mixture of MO and Span, and on the MO [24].

Temperature (ºC)	Viscosity (mm²/s)				Density (g/ml)			
	25 ºC	40 ºC	60 ºC	80 ºC	25 ºC	40 ºC	60 ºC	80 ºC
MO	17.08	9.39	5.37	3.43	0.867	0.857	0.845	0.832
MO+C60	17.38	9.86	5.46	3.56	0.867	0.856	0.845	0.832
MO+Span	17.61	9.87	5.81	3.61	0.867	0.856	0.846	0.832
MO+Span+TiO2	17.99	10.18	6.10	3.69	0.868	0.858	0.846	0.832

Raymon et al. [26] measured the viscosity of several natural ester-based NDF containing four NP (TiO_2, Cadmium sulfite, Al_2O_3 and Fe_2O_3), each at percentage concentrations of 0.05 g/l, 0.25 g/l and 0.5 g/l. They found viscosity decreases up to 20% in some of the NDF (relative to the natural ester) at 90ºC, although the viscosities of most of the NDF were unchanged. As high viscosity is one of the main problems associated with the use of ester based fluids in large transformers, these results are particularly interesting.

Figure 12 shows the kinematic viscosity of MO at 40 ºC as a function of the concentration of functionalized nanodiamonds dispersed within it [39]. The viscosity increased slowly with increasing NP concentration at concentrations below 1 % wt, but much more rapidly at higher concentrations. This increase with increasing NP concentration is in agreement with [24]. However, decreases in viscosity with increasing NP concentration have been reported for other NP/base fluid combinations [85]. However, it should be noted that NDF containing NP concentrations as high as 2.5 wt % would probably not be suitable for transformers, because their dielectric properties would probably be much inferior to those of MO.
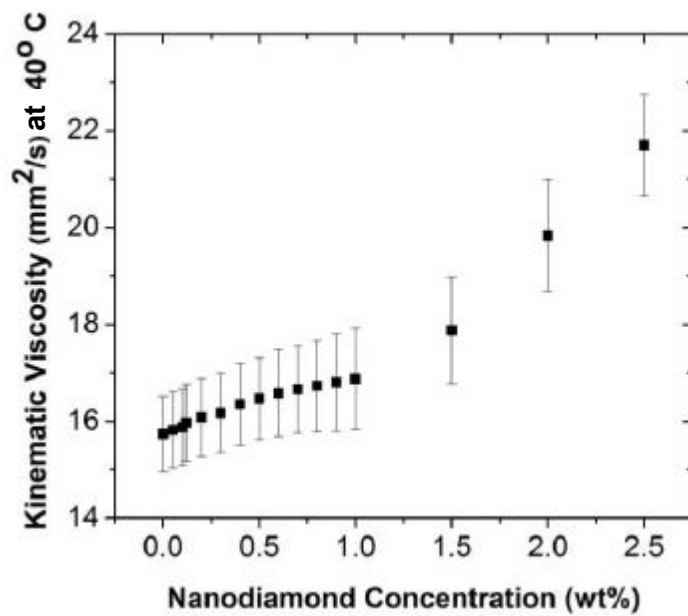

Figure 12.   Increase of the viscosity of MO-based NDT with increasing concentration of functionalized nanodiamonds [39].

7. Future Developments
Although some of the results that have been obtained for various NDF are promising, the development of these products is still at an early stage, and the prospect of using them in oil-filled transformers is still remote. It is suggested that future work be directed as follows:

   • Recently-reported NDF nearly always have short life-times (a few days to several months). Manufacturing techniques for NDF with long-term stability should therefore be developed, e.g., by searching for new surfactants and dispersion techniques.

   • Most published papers on NDF deal with their behaviour at ambient temperature. They should also be tested at transformer operating temperatures to ensure that no NP agglomeration or precipitation occurs.

   • Although some work has been done on the interaction between NDF and transformer solid insulation, the interaction between NDF and Kraft paper or pressboard in particular should be investigated in detail.

   • The results obtained by different authors often differ greatly, even for NDF with the same base and the same NP at the same concentrations. These discrepancies could be due to differences between

manufacturing methods, or between testing procedures. Widely-accepted standards should be published recommending detailed manufacturing and testing procedures.

- Finally, the behaviour of model transformers insulated with NDF needs to be closely investigated before NDF can be widely adopted in industrial transformers. Important questions include the effect of NP on transformer diagnostic techniques, e.g., dissolved gas analysis, capacitance and tan delta measurements, and how transformer field treatments, e.g., regeneration of oil and field drying, alter the performance of NDF.

Acknowledgement

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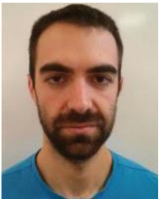
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