

Received 25 February 2024, accepted 10 May 2024, date of publication 14 May 2024, date of current version 4 June 2024.

Digital Object Identifier 10.1109/ACCESS.2024.3400986

TOPICAL REVIEW

# A Review on Electrical Breakdown of Natural Ester-Based Nanofluids Under Different Voltage Waveforms

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This work was supported by King Saud University, Riyadh, Saudi Arabia, through the Researchers Supporting Project RSPD2024R258.

**ABSTRACT** Environmental, safety, and availability constraints of mineral oils have led transformer manufacturers and researchers to turn to alternative liquid dielectrics to mineral oils. Thus, many works have focused on esters and more particularly on natural esters (NEs); commonly called vegetable oils; in recent decades. Besides their advantages (biodegradability, non-toxicity, recyclability, thermal stability, and safety) compared to mineral oils, natural esters have certain disadvantages including their high viscosity and their oxidation instability. Both of these properties can be improved by adding specific additives and nanoparticles. This paper is mainly focused on review of the breakdown voltage of natural ester-based nanofluids, one of the most significant parameters when selecting insulating fluids for high voltage devices and in particular for power transformers. Firstly, we take stock of the history of the introduction of fluids/oils in the field of electrical insulation and the return of vegetable oils in the transformer industry. Then, we present the main characteristics of vegetable oils and the different products based on vegetable oils currently on the market as well as those that are currently the subject of research and plants from which they come. The methods and techniques for the preparation/elaboration of these nanofluids and their stability are then exposed. Finally, we present the different results reported in the literature on the electrical breakdown of various types of natural esters when adding nanoparticles (NPs) issued from different materials (conducting, semiconducting, and insulating), under AC, DC and lightning impulse voltages, and discuss the involved mechanisms in the breakdown of nanofluids.

**INDEX TERMS** Power transformers, natural esters, nanoparticles, vegetable oils, different voltage waveforms.

## I. INTRODUCTION

Transformers are strategic components of electrical networks. The reliability of these latter strongly depends on the performances of these components and more particularly on the insulation system used therein. Different types of

The associate editor coordinating the review of this manuscript and approving it for publication was Ye Zhou<sup>1</sup>.

transformers are used, starting from the center of production of the electrical energy (i.e., the electrical plant) until the distribution while passing by its transport. Depending on their power and voltage levels, transformers can be dry-type or oil-filled. Most power transformers are oil-filled transformers. They are usually installed at the ends of transmission lines (i.e. at the output of the power plant and the arrival at distribution substations). Their insulation system consists

of a dielectric liquid (oil) and solid insulating (generally paper/pressboard). The dielectric liquid serves as an insulant and impregnator for solid insulating, a coolant for windings, and a protective barrier for the transformer core; the gases and decomposition products it contains also serve as indicators for diagnosing the condition of the transformer.

Different types of fluids have been used in transformers. However, the most used fluids for more than a century are mineral oils and more particularly the naphthenic ones because of their viscosity which is much lower than that of the paraffinic ones. Mineral oils have excellent dielectric and thermal properties, good oxidation stability, and low viscosity. They therefore meet the conditions required for performing the insulation and coolant functions in high voltage devices especially in liquid-filled power transformers. However, despite their interesting properties, these products are considered as dangerous for the environment due to their non-biodegradability and high flammability (because of their low flash point); the biodegradability of typical mineral oils is not more than 30 % for 28 days [1], so it takes a long time to decompose in the soil. Moreover, they are derived from petroleum products and therefore non-renewable materials. Leaks or spills of these fluids in the event of transformer failure or its explosion during operation can contaminate soil and water (groundwater) and turn into an important threat to humans and the ecosystem [2], [3]. These negative properties /characteristics of mineral oils motivated researchers and manufacturers to look for alternative sources for insulating fluids/oils fulfilling certain health and environmental criteria.

Due to their dielectric, physical, and chemical properties comparable, or even better for some, to those of mineral oils, natural esters (NEs), as they derive from plants, appear as an innovative alternative [4], [5], [6], [7], [8], [9], [10], [11], [12]. Numerous studies have been carried out on the performances of NEs over the past thirty years. It appears from the results reported in the literature that NEs are highly biodegradable green environmentally friendly liquid insulating material and non-toxic [13]. In addition, NEs are considered safe for transformers due to their higher flash point ( $> 300^{\circ}\text{C}$ ) and fire point (ranging from  $316$  to  $360^{\circ}\text{C}$ ) than that of mineral oils (about  $165^{\circ}\text{C}$ ) [14], [15]. The ability of NEs to absorb significantly more moisture than mineral oils makes it possible to slow down the aging of paper and thence the oil-paper insulation system; and therefore to extend the lifespan of the transformer [16], [17]. However, vegetable oils also have limitations /disadvantages, in particular, a higher viscosity and lower oxidation stability than those of mineral oils, which restricts their use in breathable transformers; this is why their application was initially limited to sealed transformers. Oxidation instability is indeed an obstacle to the use of NEs in power transformers, in particular NEs containing unsaturated fatty acids, due to their contact with the air during cooling. The oxidation stability of NEs can be improved by adding an oxidizing inhibitor (antioxidants) such as Butylated hydroxytoluene (BHT) or the tertiary butyl

hydroxylquinone (TBHQ) [18], [19], [20], [21], [22], [23], [24], [25]. However, the amount (doping concentration) of antioxidants to be added must be optimal so as not to affect the dielectric properties of the liquid [18], [19], [20], [21], [22], [23], [24], [25], [26], [27], [28], [29]; the recommended limit value is of 0.3 wt%. Otherwise, the electrical conductivity and dielectric loss of the oil will dramatically increase. It is also reported that the use of blends of vegetable oils with mineral oils would also improve the oxidation stability of vegetable oils [30], [31], [32], [33].

Even though vegetable oils are biodegradable, non-toxic, recyclable, stable thermally, renewable, and not listed as hazardous products, their high viscosity and low oxidation stability constitute major concerns for possible use in breathable power transformers. As previously indicated, the oxidation problem can be solved by adding an oxidizing inhibitor or by mixing them with mineral oils. However, there remains the thermal conductivity problem due to their high viscosity (i.e., the cooling of devices).

One of the solutions to solve this problem would be the addition of NPs to the insulating liquids [34], [35], [36], [37], [38], [39], [40]. This is where the concept of “nanofluids” was born. It was first introduced at the end of the 20th century by Choi and Eastman [34] who noticed that the dispersion of different metal oxide nanoparticles in a base liquid improves the heat transfer rate of this latter. Indeed, the dispersion of nanoparticles in base fluids affects various properties including thermal conduction [40]. By dispersing iron oxide ( $\text{Fe}_3\text{O}_4$ ) nanoparticles in water, a significant enhancement of thermal property and therefore the cooling property of this fluid has been observed [40]. This property is at the origin of the development of nanofluids (NFs). These have indeed better thermal diffusivity, thermal conductivity, and convective coefficient of heat transfer than the base fluids. Nowadays, NFs are used in several fields, like transportation (the cooling systems of heavy power machines and heat generation parts of vehicles); nuclear cooling systems; reducing pollution and heating buildings; space stations and aircraft; electronics (chips, electronic circuitry components); solar absorption for heat-transfer performance, etc. These NFs’ thermal properties strongly depend on the nanoparticle shape and size, concentration of nanoparticles and surfactants, the type of nanoparticles (nature and structure), method of nanofluid preparation, temperature, etc [34], [35], [36], [37], [38], [39], [40], [41].

The thermal capacity of NFs has attracted the attention of specialists in high voltage insulation and more particularly of manufacturers and operators of transformers. And for about thirty years, intense research has been performed on the physicochemical properties of nanofluids and their dielectric strength [1], [2], [3], [42], [43], [44], [45], [46], [47], [48], [49], [50]. So, it appears from the reported results in the literature that the characteristics of nanoparticles influence not only the thermal properties as mentioned above but also the dielectric properties of the nanofluid. Indeed, for a given

concentration of nanoparticles (NPs), the smaller the NPs, the higher the interfacial areas (surface contacts) between the nanoparticles and the host insulating fluid, and the higher the dielectric strength of the nanofluid will be. This improvement of the dielectric strength also depends upon the kind of nanoparticles (conductive, semiconducting, or insulating) and hosting liquid.

This paper is mainly focused on the breakdown voltage of natural esters-based nanofluids, one of the most significant parameter when selecting insulating fluids for high voltage devices and in particular for power transformers. Firstly, we take stock of the history of the introduction of fluids/oils in the field of electrical insulation and the return of vegetable oils in the transformer industry. Then, we present the main characteristics of vegetable oils and the different products based on vegetable oils currently on the market as well as those which are currently the subject of research and plants from which they come. Then we present the methods and techniques for the preparation of these nanofluids and their stability. Finally, we present the state of knowledge on the electrical breakdown of various types of natural esters - based nanofluids, under AC, DC and lightning impulse voltages; and discuss the involving mechanisms in breakdown of nanofluids.

## II. HISTORY OF THE INTRODUCTION OF DIFFERENT INSULATING LIQUIDS INTO TRANSFORMERS

The idea of using oils (dielectric liquids) for electrical insulation dates back more than a century and a half. These are vegetable oils (natural products), which were the first to be tested as a first intention to see their effects on the oxidation of electrical windings. Indeed, in 1854, C. A. Abdy [51] showed that immersing the windings in turpentine protected them from direct oxidation and thereby increased their dielectric strength and their lifespan [52]; the turpentine being obtained by the distillation of resin harvested from living trees, mainly pines [52]. At this time, castor oil, a non-edible product obtained from the seeds of the castor oil plant “*Ricinus Communis*” has been also used for the impregnation of electrical machine windings and capacitors.

In 1891, petroleum oil was used experimentally in transformer insulation [53] to replace air. It was only at the beginning of the 20th century, faced with the multiplication of electrical energy distribution networks and the increase in installed powers, that air-insulated transformers (known as “dry”) became enormous (in terms of size view). They were then gradually replaced by transformers filled with mineral oil; known as “submerged” or “oil-filled transformers”; mineral oil serving as electrical insulation and heat transfer fluid ensuring heat transfer in high voltage power transformers.

Mineral oils are derived from petroleum-based resources. Their physicochemical properties depend on the original crude oil from which they are issued. Based on their viscosity, mineral oils are grouped into naphthenic and paraffinic. Paraffinic oils have the disadvantage of freezing quickly and

preventing the operation of transformers in extreme cold. And it is because of their low viscosity, compared to that of paraffinic oils, that naphthenic oils are the most used ones in oil-filled power transformer since a century [54], [55].

One of the disadvantages of mineral oils is that they have a low flash point (low fire resistance) and therefore synthetic liquids such as polychlorobiphenyls (PCBs) were developed. So, mineral transformer oils were partially replaced by PCBs between the 1930s and 1970s. In the 1970s, their use was banned because of their toxicity and new synthetic liquids with low flammability (i.e., with high fire points), such as silicone oils or synthetic esters were developed. These new fluids make it possible to avoid electrical transformer fires [56], [57], [58], [59], [60].

Silicone oils (polydimethylsiloxane or PDMS) have very good chemical stability, which means good resistance to aging. In addition, they have a flash point almost twice as high as mineral oils, around 300°C instead of 150°C, which reduces the risk of explosion of power transformers [54], [61]. However, silicone oils do not provide the same level of dielectric insulation as mineral oils for large volumes. Moreover, high-temperature viscosity is higher than other oils; they expand more with temperature than mineral oil. They are certainly inert, but difficult to biodegrade. Also, the cost of silicone oils is also significantly higher than mineral oil (by about a factor of eight) [55]. It is for these different reasons that the use of silicon oils remains limited to small transformers (distribution and traction).

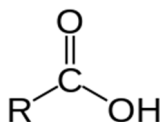
Synthetic esters, also called pentaerythritol esters or organic esters, are alternatives to mineral oils that can pass tests for immediate biodegradability tests [61], [62], [63], [64], [65]. Pentaerythritol esters are the most commonly used synthetic esters in transformers. They can absorb much more water than mineral oils, without this having a significant effect on their dielectric strength. This ability to absorb water can help preserve transformer-insulating papers and slow down their aging. These products have good dielectric strength and a fire point twice that of mineral oils. They are also more expensive than mineral oils and were not introduced until the early 80s. The use of synthetic esters remains also limited to small transformers; distribution and traction; because of their much higher cost.

Since the early 1990s, vegetable oils have made a comeback in the electrical energy sector. However, instead of turpentine or castor oil as was the case in the 1850s, today's vegetable oils come from various other plants (seeds) such as sunflower, soybean, rapeseed (or canola), corn, peanut, coconut, avocado, sesame, grape seeds, etc [4], [5], [6], [7], [8], [9], [10], [11], [12], [13], [14], [15]. Vegetable oils are also called natural or vegetable esters (as opposed to synthetic or organic esters) because they are synthesized naturally by any living organism. Their introduction in transformers results from both national and international political decisions taken in recent decades and which encourage manufacturers to increasingly develop and use products with low environmental impact and more particularly to

replace mineral oils which are liquids of fossil origin. So, many investigators have been interested in the development and characterization of these eco-friendly products for the last thirty years. Numerous studies have been achieved on vegetable-based oils to check the required criteria for their possible use in high voltage power transformers and their performance compared to mineral and synthetic oils. As indicated above, vegetable insulating oils have a very high fire point (above 300°C) and thence hardly flammable. Like synthetic esters, vegetable oils can absorb much more water than mineral oils, without this having a noticeable effect on their dielectric strength/properties [66].

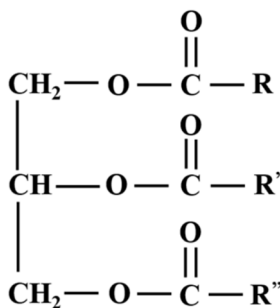
### III. VEGETABLE OILS (OR NATURAL ESTERS) AND THEIR MAIN CHARACTERISTICS

Vegetable ester oils consist of triglycerides (R-COO-R'), naturally synthesized by esterification of glycerol - a tri-alcohol, with saturated or unsaturated fatty acids [67]. The configurations of fatty acids depend on the number of hydrogen atoms present on the carbons. The fatty acids are made up of linear hydrocarbon chains ended by a COOH carboxylic acid function as shown in Figure. 1 [68]. This forms a more or less long chain depending on the type of fatty acid [68], [69].



**FIGURE 1.** COOH carboxylic acid function, R refers to the rest of the molecule to which this group is attached [68].

The unsaturated fatty acids are made up of one or more carbon-carbon double bonds (C=C), enabling them to be classified into two categories: Monounsaturated fatty acids, i.e. containing a single double bond, and fatty acids polyunsaturated compounds consisting of several carbon-carbon double bonds (C=C) as shown in Figure 2. The difference between these two types of fatty acids results from their molecular structure, the number 3 or 6 corresponds to the position of the first unsaturation or double bond (C=C). They



**FIGURE 2.** Structure of natural esters. R, R' and R'' refer to the different alkyl groups with a major variation in the number of C=C double bonds [69].

will both provide health benefits and are a great source of energy [68]. Unlike unsaturated fatty acids, saturated fatty acids have only single carbonic bonds, i.e. all the carbons are connected to hydrogen atoms. Saturated fatty acids are generally in solid form at room temperature [68]. The fatty acid components of triglycerides play an important role in determining the physicochemical properties of vegetable oil.

Vegetable insulating oils are bio-sourced products, easily biodegradable and non-toxic [4], [5], [6], [7], [8], [9], [10], [11], [12], [13], [14], [15], [69]. They are recommended in certain countries when there is a risk of accidental contact with the water table. The life cycle assessment of vegetable transformer oil indicates a much lower environmental impact than that of mineral oil [70], [71], [72], [73], [74]. They generally have a higher pour point than mineral oils (between -20 and -32°C depending on the product). The chemical structure of vegetable insulating oils gives them a great capacity to absorb water, unlike mineral oils [68]. This property allows them to dry out the insulating papers of transformers, and extend their lifespan. Vegetable insulating oils are less likely to generate sludge and deposits as they age, which is an advantage over mineral oils. They are sensitive to excess oxygen: to avoid their long-term degradation, they must only be used in hermetic transformers.

The main products based on vegetable oils, developed the last thirty years, and currently available on the transformer market are FR3 <sup>®</sup>, BIOTEMP <sup>®</sup>, and MIDEL eN 1204. These are extracted from sunflower, rapeseed, canola, and soybean [10], [75], [76], [77], [78], [79].

FR3 Cargill (Envirotemp<sup>®</sup>FR3<sup>™</sup>) fluid is a vegetable natural ester derived from more than 95% rapeseed oil, a renewable source. This natural ester serves also as a coolant for use in distribution and power class transformers where its fire safety, environmental, electrical, and chemical properties are advantageous. It is a K-class product with a fire point of 360°C, non-toxic and non-hazardous in soil and water, readily biodegradable, contains no petroleum, halogens, silicones, or sulfurs and is recyclable [67], [75], [76]. It is a more efficient, more reliable, and more environmentally friendly dielectric fluid than mineral dielectric oils. FR3 fluid absorbs free water and essentially wicks it away from the insulation paper. FR3 fluid has 10 times the water saturation level of mineral oil. This results in extending the insulation life 5-8 times longer than mineral oil. The fluid is also used in retro-fill applications for transformers and other fluid-filled distribution and power equipment. FR3 Cargill fluid is the most widely used [67], [75], [76].

MIDEL eN 1204 and MIDEL eN 1215 are two sustainably sourced from rapeseed and canola crops, respectively [10]. These natural esters are safer than mineral oil, non-toxic, and readily biodegradable, avoiding environmental damage should leakage occur and enabling reductions in containment measures. They offer superior moisture tolerance and have the potential to increase the lifetime of cellulose-based solid insulation, which in turn can extend transformer life.



The main difference is in the pour point which is  $-31^{\circ}\text{C}$  and  $-18^{\circ}\text{C}$  for MIDEL 1204 and 1215, respectively.

BIOTEMP, produced by ABB, Inc., is a natural ester-based dielectric insulating fluid used in transformers. It is made from high oleic sunflower or safflower seed oil [77], [78], [79]. This product is biodegradable and “less flammable”; its fire point is above  $300^{\circ}\text{C}$ . Its heat capacity and heat transfer properties are superior to those of conventional mineral oils.

Unfortunately, most current vegetable oils available on the transformer market are derived from seeds of food crops which, over time, risks generating new humanitarian, ethical and economic problems. Therefore, the prices of some of these feedstock will be subject to significant fluctuations caused by the food market. Their use for the production of transformer oils would further increase the cost of production and therefore the prices of transformer oils and would contribute to food crises. It is therefore needed to divert our attention to non-food materials so that the searches of new material alternatives for replacing mineral oil do not interfere with foodstuffs.

This verdict led researchers/manufacturers to move towards non-edible resources. Since investigations on these latter (non-edible resources) have been engaged, and their number increases from year to year. Among these types of oils, one can cite: Jatropha methyl ester oil - JMEO [80], [81], palm kernel oil [82], [83], terminalia catappa oil [84], Karanja oil [85], castor oil [86] and Neem oil [87], [88]. Although these oils meet the criteria required by standards in terms of environmental protection and toxicity, they have some weak points. Among these, is their pour point especially for transformers operating in cold regions. The pour point of vegetable oils does not drop below  $-30^{\circ}\text{C}$  even when depressants are added, oil could freeze at subzero degrees in the absence of additives [89].

Some of these non-edible vegetable oils have been patented for use in transformers. Among these, is Jatropha methyl ester (oils) – JMEO which is derived from jatropha curcas seeds [2]. Jatropha curcas oil presents several advantages that recommend both its production and usage over those of other vegetable oils as crude palm oil and rapeseed oil. Indeed, jatropha curcas may be grown on marginal or degraded soils avoiding thus the need to utilize those more fertile soils being used to grow the staple crops; and it readily grows in areas where annual rainfall levels are significantly lower than those required by other species such as palm oil, rapeseeds oil, sunflower oil, soybeans oil, corn oil and others. Jatropha curcas plants can grow on all soil types, even on barren soil. It is widely distributed in the wild and abundant in both tropical and subtropical regions (America, Asia, Africa, Pacific, Australia) [2], [90], [91], [92], [93], [94], [95].

The viscosity of crude jatropha curcas oil (CJO) is approximately 29-33 cSt at  $40^{\circ}\text{C}$ . This value is much higher than that of mineral oil, which is less than 12 cSt at  $40^{\circ}\text{C}$ . But this value is still appropriate according to the IEEE C.57.147 standard which recommends values below 50 cSt at  $40^{\circ}\text{C}$ . On the other hand, the acid number of crude jatropha oil (CJO) is

in the range 32-33 mg KOH/g [2]. This value is much higher than that required by the IEEE C.57.147 standard which is  $\leq 0.06$  mg KOH/g. Given its viscosity and its acidity index, jatropha curcas oil does not meet the IEEE C.57.147 standard, hence the need to subject it to an esterification process to reduce its viscosity and its acidity index. This process aims to convert free fatty acids (FFA) and triglycerides into methyl esters and glycerol.

Esterification is a process of breaking (cutting) the C-O bonds between triglycerides and methanol on the carbonyl carbon. Through the esterification process, one obtains the methyl ester called “JMEO (Jatropha Methyl ester oil)”. JMEO has been patented in 2010 [96] and the results of investigations on this ester has been reported [2], [30], [96], [97].

Research is underway around the world on other vegetable oils made from non-edible plants/seeds to qualify and patent them so that they can be used on a large scale.

#### IV. NATURAL ESTER-BASED NANOFLUIDS

##### A. PREPARATION OF NANOFLUIDS' SAMPLES

Nanofluid preparation is a fundamental step as their physicochemical properties, in particular thermal and dielectric, depend on it [40]. There are two methods for preparing nanofluids: the one-step method and the two-step method.

The one-step method consists of synthesizing and dispersing simultaneously the nanoparticles in the base fluid [2], [30], [40], [50], [98], [99], [100], [101]; it does not require drying, storage, or transportation of nanofluids. This method allows controlling the size and shape of NPs during the synthesis process while minimizing the agglomeration and avoiding the use of additional stabilizers, and therefore to obtain stable NFs [101]. This method is indicated for the weak concentration of NPs and small volume of NFs [102], [103]. Moreover, it is costly and unsuitable for large-volume production [99].

For the large-scale production of nanofluids, particularly for transformer applications, the two-step method is widely used [98], [99]. In this method, the nanoparticles are first dispersed in the base fluid using a magnetic stirrer or other homogenizing technique as depicted in Figure 3. To reduce the interaction forces between the nanoparticles and the high surface energies of NPs which can affect the stability of NF, a surfactant is added to the solution; the surfactant also allows a better immersion of the nanoparticles. The most widely

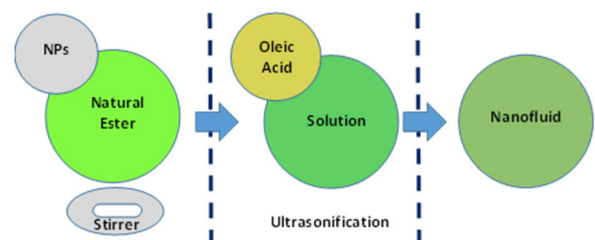


FIGURE 3. Diagram of two-step method for preparation of nanofluids (NFs).

used surfactant is oleic acid. Long-chain hydrocarbons such as sorbitan esters, hexadecyl trimethyl ammonium bromide (CTAB), and sodium dodecyl sulfate (SDS) are also used, but rarely [99], [100]. Homogeneity and stability are controlled once the nanofluid is prepared. Note that the excess amount of surfactant can affect the stability of nano-based oil and uniform dispersion of nanoparticles due to the formation of a double chain of surfactant around nanoparticles.

Regarding dispersion, the most used technique is ultra-sonification; it is well known to be very efficient and reliable for the preparation of nanofluids [104], [105], [106], [107]. Ultra-sonification involves applying high-frequency sound waves to cause pressure changes in the fluid. This makes it possible to break the clusters of nanoparticles into tiny nanoparticles and thus disperse them in the medium [108]. Another influential parameter during the ultra-sonification process is its application duration; this latter affects the thermal properties of the nanofluid. It was reported that the thermal conductivity of nanofluid increases with the ultra-sonication time [109]. The higher the ultra-sonication power, the smaller the size of the aggregates will be, and the higher the zeta potential the better the particle dispersion will be [110]. Note that both the average cluster size and ultra-sonication time duration zeta potential [111].

The zeta potential ( $\zeta$ ) is an indicator of the stability of a dispersion. The stability of NF is all the better as the dispersion of the nanoparticles is homogeneous; it directly impacts the thermal transfer of NF; settling, aggregation, and clustering of nanoparticles within the nanofluids are a threat to the thermal conductivity of NF. The zeta potential is expressed in millivolts and is generally between -200 mV and +200 mV; the sign (positive or negative) of the dispersion does not matter for stability. Zeta potential is not a direct measure of the stability of a dispersion, but it provides a good prediction of stability. This is an easy and quick measure to carry out.

## B. BREAKDOWN

Studies on the dielectric strength of natural esters-based nanofluids (NFs), under different voltage waveforms (AC, DC, and LI - lightning impulse) have been carried out since the early 1990s. They relate to the effect of conductive, semiconducting, and insulating nanoparticles (NPs) and their concentration on the breakdown voltage of these nanofluids; the sizes of NPs range from some nm to a hundred nm. The most investigated NPs are metal oxides such as titanium  $\text{TiO}_2$ , magnetite  $\text{Fe}_3\text{O}_4$ , alumina  $\text{Al}_2\text{O}_3$ , silicone oxide  $\text{SiO}_2$ , and zinc oxide  $\text{ZnO}$ . Other types of nanoparticles such as fullerene ( $\text{C}_{60}$ ), magnesium oxide ( $\text{MgO}$ ), Zirconia ( $\text{ZrO}_2$ ), Graphene ( $\text{C}_n$ ), graphene oxide (GO), and copper oxide ( $\text{CuO}$ ), have been also considered.

The results reported in the literature on the impact of some types of NPs on the dielectric strength of natural esters are controversial. Some NPs have a beneficial effect while others, on the contrary, degrade the dielectric strength of the base liquid depending on the type of NPs, their sizes and concentrations; and the voltage waveform.

### 1) AC BREAKDOWN VOLTAGE

The knowledge of the dielectric strength of any insulating material is fundamental for the sizing of electrical devices required to operate under AC voltage. Therefore, measuring the AC breakdown voltage of nanofluids is essential for their possible use in liquid-filled apparatus, especially in transformers.

In the following, we present a synthesis of results reported in the literature as well as our experimental findings performed according to the IEC 60156 standard (i.e., using a spherical electrode configuration of 12.5 mm diameter, spaced 2.50 mm apart) or to the equivalent standards.

As indicated above, the impact of NPs on the dielectric strength of a given liquid depends on their type (material nature), shape, size, and concentration.

By considering conductive ( $\text{Fe}_3\text{O}_4$ ) and two insulating ( $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$ ) nanoparticles, Usama and Beroual did not observe a significant influence on the AC breakdown voltage (AC-BDV) of MIDEAL 1204 natural ester [112]. The best improvement is about 7%; it is obtained with  $\text{Fe}_3\text{O}_4$  of 50 nm size at a concentration of 0.4 g/L and  $\text{Al}_2\text{O}_3$  of 13 nm size at a concentration of 0.05 g/L; it is too weak compared with the results obtained with mineral oil or synthetic ester MIDEAL 7131 [45], [49]. Indeed, with  $\text{Fe}_3\text{O}_4$  (50 nm size) nanoparticles at a concentration of 0.4 g/L, the improvement of the AC-BDV can exceed twice that of pure mineral oil alone; and with a concentration of 0.05 g/L of  $\text{Al}_2\text{O}_3$  (13 nm size) NPs, the AC-BDV can be enhanced by 72% with respect to synthetic ester MIDEAL 7131 [49]. Sometimes, the addition of nanoparticles to natural ester has a negative effect on the dielectric strength; it degrades the AC-BDV as it is the case of  $\text{Fe}_3\text{O}_4$  and  $\text{Al}_2\text{O}_3$  of 50 nm size at a concentration of 0.05 g/L where the reduction is of about 16% and 10%, respectively. In the case of  $\text{SiO}_2$  of 50 nm size at a concentration of 0.05 g/L, the degradation is of about 15%. Table 1 gives the mean AC-BDV of MIDEAL 1204 natural-based nanofluids. With  $\text{Al}_2\text{O}_3$  nanoparticles of 50 nm size, the breakdown voltage of NFs reduces by about 4% for 0.2 g/L concentration and decreases by about 6% for 0.3 g/L concentration. While it is slightly improved at 0.3 g/L (by about 6%); with 0.4 g/L, the improvement is negligible. For the same kind of nanoparticles ( $\text{Al}_2\text{O}_3$ ) with 13 nm size, the enhancement is insignificant (1.6%) at a concentration of 0.2 g/L; and the breakdown voltage reduces at 0.3 and 0.4 g/L (by about 2%). Concerning  $\text{SiO}_2$  NPs of 50 nm size, the breakdown voltage of NF reduces by about 15% at 0.05 g/L concentration, compared to the base oil. Then, it increases by about 5.3% and 5.8%, at 0.2 g/L and 0.3 g/L concentrations, respectively; it decreases by about 2% at 0.4 g/L. Therefore, the variation is irregular; it is jagged. Therefore, adding these three types of NPs ( $\text{Fe}_3\text{O}_4$ ,  $\text{Al}_2\text{O}_3$ , and  $\text{SiO}_2$ ) has no spectacular effect on the AC-BDV of MIDEAL 1204 natural ester; the effect can be even negative. Therefore, from the point of view of breakdown voltage, MIDEAL 1204 natural ester oil based- $\text{Fe}_3\text{O}_4$ ,  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  nanofluids are not so interesting as it is the case with mineral oil and synthetic ester with the same

**TABLE 1. AC mean breakdown voltages of different natural ester-based nanofluids [112].**

	Natural ester oil	Fe <sub>3</sub> O <sub>4</sub> (50nm)	Al <sub>2</sub> O <sub>3</sub> (50nm)	Al <sub>2</sub> O <sub>3</sub> (13nm)	SiO <sub>2</sub> (50nm)
NE / 0.05 (g/L) NF					
BDV (kV)	68.77	57.53	61.53	73.93	58.20
Increment (%)	-	-16.34	-10.53	7.56	-15.37
NE / 0.2 (g/L) NF					
BDV (kV)	68.77	66.90	66.10	69.87	72.23
Increment (%)	-	-2.72	-3.88	1.59	5.03
NE / 0.3 (g/L) NF					
BDV (kV)	68.77	69.53	73.00	67.33	72.73
Increment (%)	-	1.11	6.15	-2.09	5.76
NE / 0.4 (g/L) NF					
BDV (kV)	68.77	73.63	69.07	67.30	67.13
Increment (%)	-	7.07	0.44	-2.14	-2.38

nanoparticles. It would also be difficult to envisage the use of this type of nanofluids, at least based on this ester. This would bring no added value compared to vegetable oil alone.

Similar results have been reported by Peppas et al. [45], [46], [113] regarding the effects of Fe<sub>3</sub>O<sub>4</sub> and SiO<sub>2</sub> nanoparticles on the AC-BDV of FR3 (Envirotemp<sup>TM</sup>) natural ester oil. They observed that the addition of 0.008% wt./wt. of commercial Fe<sub>3</sub>O<sub>4</sub> powder or 0.012% wt./wt. of oleate-coated colloidal Fe<sub>3</sub>O<sub>4</sub> increases the AC-BDV by 20% with respect to the base oil (natural ester oil). With further addition of Fe<sub>3</sub>O<sub>4</sub> NPs the AC-BDV drastically drops well below the breakdown voltages of the base oil. They attributed this to the decreasing inter-particle distance of the Fe<sub>3</sub>O<sub>4</sub>, NPs which start forming conductive paths above a threshold value. Also, according to these authors, the AC-BDV of NFs with SiO<sub>2</sub> NPs is lower than that of natural ester.

The impact of Fe<sub>3</sub>O<sub>4</sub> (conductive) and TiO<sub>2</sub> (semi-conducting) NPs on the AC-BDV of a highly refined, bleached, deodorized palm oil (RBDPO) has been also investigated by Makmud et al. [47]. By considering concentrations of 0.01, 0.1, and 1.0 g/L, they observed a beneficial effect of TiO<sub>2</sub> NPs; the AC-BDV increases by about 16%, 48%, and 68 %, respectively. While, with Fe<sub>3</sub>O<sub>4</sub> NPs, the AC-BDV increases only for the low concentration, i.e., at 0.01 g/L; let be a rate of about 22%. For the other two concentrations (0.1 and 1.0 g/L), the AC-BDV decreases by about 25% and 58%, respectively.

Mohamad et al. [114] measured the AC-BDV of palm fatty acid ester (PFAE) based nanofluids with three types of nanoparticles at a concentration of 0.01 g/l, namely Fe<sub>3</sub>O<sub>4</sub>, TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> nanoparticles, the NPs sizes varying between 15 to 20 nm. They observed that the PFAE oil-based Fe<sub>3</sub>O<sub>4</sub> nanofluid shows exceptional dielectric performance compared to other NF samples. The AC-BDV increases by 43%, 29% and 34% with Fe<sub>3</sub>O<sub>4</sub>, TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, respectively. To explain these results, the authors highlight the mechanisms put forward by Yuzhen et al. [43].

A study of the influence of conductive (Fe<sub>3</sub>O<sub>4</sub>), semi-conductive (CuO) and insulating (Al<sub>2</sub>O<sub>3</sub>) NPs without and with a surfactant, on the AC-BDV of Refined, Bleached and Deodorized Palm Oil (RBDPO) Olein and Coconut Oil (CO), has been carried out by Mohamad et al. [102]; the surfactant being Cetyl Trimethyl Ammonium Bromide (CTAB) and the sizes of NPs are 50 – 100 nm for Fe<sub>3</sub>O<sub>4</sub>, <50 nm for CuO and 40 nm for Al<sub>2</sub>O<sub>3</sub>. By varying the concentration of NPs from 0.001% to 0.05%, they observed an improvement of the average AC-BDV of RBDPO and CO only with Al<sub>2</sub>O<sub>3</sub> NPs. Using Weibull distribution analysis, they found that the AC-BDV at 1% probability for RBDPO and CO is enhanced when adding Fe<sub>3</sub>O<sub>4</sub>, CuO and Al<sub>2</sub>O<sub>3</sub> NPs at certain volume concentrations. The highest improvement is obtained with Al<sub>2</sub>O<sub>3</sub> for both RBDPO and CO; it can reach 52%. However, the effect of CBTA on the AC-BDV of both liquids is not clear.

In a work performed by Li et al. [115], it was observed that the addition of Fe<sub>3</sub>O<sub>4</sub> and oleic acid increases the average AC-BDV of rapeseed oil by 20%. This enhancement is attributed to the trapping of free electrons by the polarized nanoparticles, and thus to prevent streamer propagation [116].

Zhong et al. [117] observed that the introduction of TiO<sub>2</sub> semi-conductive nanoparticles in natural ester FR3 (a product derived from rapeseed), at a concentration of 0.3 g/L enhances the AC-BDV by 31%. According to these authors, this could be due to the electron trapping and un-trapping processes in the shallow traps. A similar result has been reported by Olmo et al. [118]. They noticed that the addition of TiO<sub>2</sub> NPs (10-20 nm) improves the AC-BDV of a natural ester (NE) derived from sunflower seeds by 33.2% at a concentration of 0.5 g/L, while with the other concentrations, the enhancements are 25.8%, 30.4%, 21.6%, and 7.6% for the concentration of 0.1, 0.2, 0.7, and 1 g/L, respectively. To explain these results, the authors advanced the same theory as that proposed by Zhong et al. [117].

Hussain et al. [119] showed that adding magnetic NPs such as iron oxide (Fe<sub>3</sub>O<sub>4</sub>), cobalt oxide (Co<sub>3</sub>O<sub>4</sub>), and iron phosphide (Fe<sub>3</sub>P) enhances the dielectric strength of NE. The Fe<sub>3</sub>P NFs give the best improvement for a concentration of 0.02 g/L; let be 31.4%. These authors also rely on the same theory as that of put forward by Zhong et al [117] to explain this enhancement.

Asse et al. [120] analyzed the impact of FeO<sub>3</sub> NPs on the AC-BDV of Palm Kernel Oil Methyl Esters (PKOME). They observed a 40% improvement in breakdown voltage for the 0.10 wt% concentration, 20% and 37.3% for the 0.15 wt% and 0.20 wt% FeO<sub>3</sub> concentrations, respectively.

Duzkaya and Beroual measured the AC-BDV of the MIDEL eN 1204 natural ester when adding ZnO nanoparticles (25 nm diameter) at 0.05 and 0.4 g/L concentrations [101]. The tests were performed according to the IEC 60156 standard in a horizontally positioned sphere-sphere electrode arrangement. They reported that the AC-BDV of natural ester-based ZnO nanofluids decreased



for 0.05 and 0.4 g/L concentrations and increased for 0.1 to 0.3 g/L concentrations. The best improvement is obtained with a concentration of 0.1 g/L ZnO; which is about 5%. Beyond a concentration of 0.4 g/L, the AC-BDV decreases. The experimental data were also analyzed using Weibull and normal distribution functions; and the risks of 1%, 10% and 50% probabilities of breakdown voltages are also computed using these distribution functions. The statistical analysis shows that the best improvement in 1% probability withstand voltage is 22.7% and it is in 0.1 g/L ZnO nanofluid. For the same probability, the enhancement of the AC-BDV is 13.2% and 11.1% at concentrations of 0.2 and 0.3 g/L, respectively. The best performance for 10% probability is also in the 0.1 g/L concentration; it is 13%. At this stage, it is very difficult to propose a mechanism that could explain this reduction.

In another work, Beroual and Duzkaya [121] examined the variation of AC breakdown voltages of the same natural ester enriched with fullerene ( $C_{60}$ ) nanoparticles for concentrations ranging from 0.05 to 0.4 g/L, the average size of NPs being 21 nm. They observed that 0.3 and 0.4 g/L  $C_{60}$  nanofluids present better performances compared to the base natural ester with an increase rate of 5.1% and 7.8%, respectively. The AC breakdown voltage of 0.05 g/L  $C_{60}$  doped nanofluid is reduced by 12.6% compared to natural ester. Decreases can be observed in the measurements of AC-BDV of natural ester-based nanofluids with low concentration nanoparticles [121]. This decrease can reach rates of 16.34%, 17.20% and 15.37%, respectively, in natural ester-based nanofluids to which  $Fe_3O_4$ ,  $Al_2O_3$  and  $SiO_2$  NPs were added at 0.05 g/L concentrations [112].

The effect of fullerene ( $C_{60}$ ) NPs on the AC-BDV of FR3 natural ester has been also investigated by Khelifa et al. [122]; the diameter of NPs varies from 4 to 8 nm. It was observed that the mean value of AC-BDV of FR3 base oil decreases when adding 0.1 g/L and increases beyond this concentration. There is an improvement of 13.08% with a concentration of 0.4 g/L. In the same work, these authors noticed that 20 nm graphene (Gr) nano-sheets reduce the AC-BDV value for a concentration of 0.5 g/L, while they increase it for concentrations of 0.1 g/L, 0.2 g/L, 0.3 g/L and 0.4 g/L compared to the base liquid. The best improvement is 12.79%; it is obtained with 0.3 g/L of Gr NPs [122].

By adding graphene oxide NPs to a soyabean-extracted natural ester, Siddique et al. [123] observed that at low concentrations, varying from 0.001 and 0.003 g/L, the AC-BDV decreases while by increasing this concentration to 0.005 g/L, the measured AC-BDV voltage increases by approximately 17 %.

Prasad and Chandrasekar [124] showed that the presence of  $SiO_2$  in the same natural ester - FR3, increases the AC breakdown voltage by 33%. The silica-coated titanium (ST  $TiO_2$ ) particles also improve the AC-BDV of Envitrafol, a natural ester extracted from rapeseed oil. Indeed, the addition of such NPs with a concentration of 0.25% results in an enhancement of more than 33% (from 60 kV for pure liquid to 80.15 kV);

the tests were performed according to IEC 60156. According to these authors, the improvement would be due to the increased shallow trap density in the liquid [116].

Khelifa et al. [131] investigated the impact of Zirconium dioxide ( $ZrO_2$ ) on FR3 natural ester by considering five concentrations of NPs ranging from 0.1 g/L to 0.5 g/L. They observed an improvement of the AC-BDV, except for 0.1 g/L and 0.5 g/L  $ZrO_2$  concentrations which degrade the dielectric strength of the base oil. They evidenced the existence of an optimal concentration at 0.4 g/L  $ZrO_2$  giving an improvement of the AC-BDV by 8.39%. These experimental findings were discussed using a trapping mechanism. By dispersing Cerium Oxide ( $CeO_2$ ) NPs in rapeseed oil at 0.02 g/L concentration and in soybean oil at 0.03 g/L concentration, Rahman et al. [126] observed an enhancement of the breakdown voltage by up 18% and 19%, respectively. These authors noticed a general trend of variation in the AC-BDV which depends on the concentration of NPs: an increase of the AC-BDV for small concentrations of NPs and a decrease beyond an optimum concentration corresponding to the maximum value of the AC-BDV; this latter was attributed to possible agglomeration of NPs.

Farade et al [127] measured the breakdown voltage of cottonseed oil (CSO)-based h-BN (hexagonal boron nitride) nanofluids at different weight percentages of NPs ranging from 0.01 wt% to 0.1 wt%. They observed an increase of the AC-BDV that reached a maximum percentage increment of 63.3% at a weight percentage of 0.1 wt%.

It appears from the different reported results that the introduction of NPs in natural esters carries risks. The AC dielectric strength of a given nanofluid can increase or decrease depending on the type of nanoparticles and their size. One cannot therefore generalize the results reported by this or that group of researchers. In some cases, nanofluids based on natural esters can be very interesting while in other cases, they can prove fatal. It is therefore necessary to be very careful before considering the introduction of a given nanofluid into a device, in particular into a transformer.

## 2) DC BREAKDOWN VOLTAGE

There is very little work on the effect of nanoparticles on the DC dielectric strength of natural esters. Among the rare works carried out under DC voltage, are those due to Usama and Beroual [128]. They investigated the impact of magnetic ( $Fe_3O_4$ ) and insulating ( $Al_2O_3$  and  $SiO_2$ ) NPs of various sizes and at different concentrations, on the DC-BDV of natural ester MIDEAL 1204 NE [128]; and compared their experimental results with the AC-BDV of the same NE with the same NPs as well as with those obtained with mineral oil (MO) and synthetic ester MIDEAL 7131 (SE).

They also analyzed the experimental results using both normal and Weibull probabilistic functions; and deduced the 1% and 50% DC-BDVs. They observed that (1) the effect of NPs on DC-BDV depends on both their type and their concentration, and (2) the improvement of DC-BDV of NE-based NFs is less important than that of the AC-BDVs of



**TABLE 2.** Average DC-BDVs of different NFs [128].

	NE oil	Fe <sub>3</sub> O <sub>4</sub> (50 nm)	Al <sub>2</sub> O <sub>3</sub> (50 nm)	Al <sub>2</sub> O <sub>3</sub> (13 nm)	SiO <sub>2</sub> (50 nm)
NE / 0.05 (g/L) NF					
BDV (kV)	56.63	56.28	46.89	55.53	53.29
Increment (%)		-0.62	17.20	-1.94	-5.90
NE / 0.2 (g/L) NF					
BDV (kV)	56.63	62.61	60.68	56.43	53.86
Increment (%)		10.56	7.15	-0.35	-4.89
NE / 0.3 (g/L) NF					
BDV (kV)	56.63	48.79	61.88	60.95	44.46
Increment (%)		-13.84	9.27	7.63	-21.49
NE/ 0.4 (g/L) NF					
BDV (kV)	56.63	52.51	47.74	60.09	36.63
Increment (%)		-7.28	-15.70	6.11	-35.32

MO and SE. However, for the concentration of 0.05 g/L, the DC-BDV decreases irrespective of the type of NP. The best enhancement is 10.6% using Fe<sub>3</sub>O<sub>4</sub> (50 nm) at 0.2 g/L under DC, whereas under AC, it is of approximately 7% using Fe<sub>3</sub>O<sub>4</sub> (50 nm) at 0.4 g/L; and the best enhancement is of 9% using Al<sub>2</sub>O<sub>3</sub> (50 nm) at 0.3 g/L under DC, whereas under AC, it is of 7% using Al<sub>2</sub>O<sub>3</sub> (13 nm) at 0.05 g/L. The highest DC-BDV improvement with Al<sub>2</sub>O<sub>3</sub> (13 nm), which is 7.6%, is observed with 0.4 g/L. However, the DC-BDV decreases irrespective of the concentration using SiO<sub>2</sub>. As far as SiO<sub>2</sub> NPs, the DC-BDV decreases whatever the concentration of SiO<sub>2</sub> (50 nm size). While the AC-BDV was lower than that of the pure oil (NE) at 0.05 g/L of SiO<sub>2</sub>; then, there is a slight improvement of about 5% at 0.2 g/L and 4% at 0.3 g/L; it somewhat decreased at 0.4 g/L. Note that whatever the type of oil used (MO, SE, or NE liquids); the best enhancement was achieved using magnetic NPs (Fe<sub>3</sub>O<sub>4</sub>). Table 2 gives the Average DC-BDVs of different NFs.

The fact that nanoparticles do not always improve the DC dielectric strength of natural ester, has been also reported by other investigators with other types of NPs. Chetty et al [129] indeed observed that the addition of 5% weight of silica aerogel (Si(OR)<sub>4</sub>) powder NPs reduces the dielectric performance of vegetable oils, and the streamer activity is less aggressive in nano-modified linseed oil than in nano-modified castor oil.

### C. LIGHTNING IMPULSE BREAKDOWN VOLTAGE

The knowledge of the lightning impulse breakdown voltage is useful to predict the behavior of oil-filled apparatus in case of lightning strikes and to size them in consequence. The lightning impulse breakdown voltage (LI-BDV) has been generally investigated under divergent electric fields and mainly in a point-plane electrode configuration. It was reported that the nanoparticles have generally a beneficial effect on the LI-BDV of esters.

So, Li et al. [48] measured the lightning impulse breakdown voltages of a vegetable insulation oil (named RDB) extracted from raw rapeseed seeds and the RDB based- Fe<sub>3</sub>O<sub>4</sub> nanofluid using a point-to-plane electrode

configuration according to IEC 60897 standards [130]; RDB is a refined, bleached and deodorized oil extracted from a myriad of oils from canola to soy to olive to sunflower to grapeseed and more. They reported that the positive average breakdown voltage of the nanofluid samples is approximately 37% greater than that of the base oil, and the negative average breakdown voltage of the nanofluid samples is approximately 12% greater than that of the base oil. They also noted that the average times to positive and negative LI-BDV in the nanofluids are, respectively, 21 and 14% greater than those in the base oil. The corresponding average velocities of streamers (discharges) are 1.25 and 1.18 km/s for positive and negative lightning impulse voltages in NFs, respectively, and 1.51 and 1.35 km/s for the base oil. They attributed these differences to the major impact of the charge relaxation time constant of NPs ( $\tau$ ) on electro-hydrodynamic (EHD) phenomena in NFs. Using the values of conductivities and permittivities of both NPs and base liquid, they computed  $\tau$ , according to the following equation [121]:

$$\tau = \left( \frac{2\varepsilon_1 + \varepsilon_2}{2\sigma_1 + \sigma_2} \right)$$

where  $\varepsilon_{1,2}$  and  $\sigma_{1,2}$  are the permittivities and conductivities of the base liquid and NP, respectively.

They found that the value they obtained is very short with respect to that of the streamer velocity. They concluded that Fe<sub>3</sub>O<sub>4</sub> NPs will significantly affect the electro-dynamics of streamer development.

When submitted to an electric field, the nanoparticles trap free electrons and become negatively charged. These negatively charged NPs are relatively heavy; their mobilities will be smaller than that of free electrons. The space charge they constitute reduces the electric field and impacts the development of the space charge at the end of a streamer which is the driving force behind the propagation of the streamer. Thus, the electrical breakdown will occur at higher voltages in NFs than in the base oils.

Beroual and Khaled [131], [132] studied the influence of magnetic (Fe<sub>3</sub>O<sub>4</sub>) and insulating (Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub>) NPs on the negative lightning impulse breakdown voltage (LI-BDV) of MIDEL 1204 natural ester. The measurements were performed using a quasi-uniform electric field with a test cell containing two brass spherical electrodes of 12.5 mm diameter; the inter-electrode interval was fixed at  $2.50 \pm 0.05$  mm. It was observed that the LI-BDV of NE is improved by the introduction of these types of NPs. In addition, for a given type of NP, there is an optimal concentration for which the  $U_{50\%}$  LI-BDV is the highest. This latter is enhanced by about 16.8% with Al<sub>2</sub>O<sub>3</sub> at a concentration of 0.05 g/L. However, the effect of conducting NPs (Fe<sub>3</sub>O<sub>4</sub>) is less important under lightning impulse voltage than under AC. Table 3 gives the  $U_{50\%}$  of different LI-BDV of different NE-based NFs.

As with synthetic ester (SE), Beroual and Khaled [131], [132] observed the existence of an optimum concentration of NPs for which there is an optimum enhancement of LI-BDV of NE. This improvement is of about 7.5% with Fe<sub>3</sub>O<sub>4</sub> at a

**TABLE 3.**  $U_{50\%}$  LI-BDV of different NE-based NFs [131].

	NE	Fe <sub>3</sub> O <sub>4</sub>	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>
NE / 0.05 (g/L) NF				
BDV (kV)	129.34	132.73	151.02	139.37
Increment (%)		2.62	16.76	7.75
NE / 0.2 (g/L) NF				
BDV (kV)	129.34	139.05	144.08	146.28
Increment (%)		7.51	11.40	13.10
NE / 0.3 (g/L) NF				
BDV (kV)	129.34	123.59	136.90	145.43
Increment (%)		-4.45	5.85	12.44
NE/ 0.4 (g/L) NF				
BDV (kV)	129.34	118.62	132.08	142.09
Increment (%)		-8.29	2.12	9.86

concentration of 0.2 g/L; 16.8% with Al<sub>2</sub>O<sub>3</sub> at a concentration of 0.05g/L; and 13.1% with SiO<sub>2</sub> at a concentration of 0.2 g/L. According to these authors, this enhancement is due to the trapping of charges (electrons) by the nanoparticles first in the vicinity of electrodes and then within the liquid volume. The accumulation of injected charges at the surface of NPs results in the decrease of the electric field in the vicinity of the electrode and thence in an increase of the initiation threshold voltage of streamers (discharges); and within the liquid volume, the nanoparticles catch the charges (electrons), reduce their numbers/amount and slowing the streamers increasing of the breakdown voltage. This process continues until the NP/host liquid interface is saturated. This saturation would explain the existence of the optimal concentration of NPs. Therefore, the properties of the nanoparticles and the liquid play a fundamental role.

Koutras et al. [133] reported that Al<sub>2</sub>O<sub>3</sub> NPs with a concentration of 0.004% w/w increase the LI-BDV of Envirotemp™FR3™ natural ester, under both positive and negative polarities by 28.5% and 1.52%, respectively. The significant improvement of the LI-BDV of NFs would be attributed to the positive charges induced by alumina NPs slowing down the positive streamer propagation. The same group examined the impact of TiO<sub>2</sub> nanoparticles on natural ester oil Envirotemp™FR3™ fluid, in concentrations ranging from 0.005%–0.040% w/v [134]. They observed an increase of 12.94% in LI BDV. The same group reported that the SiC NPs increased the LI BDV by 9.8% with 0.004% (w/w) [135]. The addition of 0.01% (w/w) of MgO NPs to natural ester leads to an enhancement of 15.15% in LI-BDV with respect to the same base oil [136].

Beroual and Duzkaya [121] measured the LI breakdown voltages (LI-BDV) of Midel eN 1204 transformer oil-based C<sub>60</sub> nanofluids with concentrations of C<sub>60</sub> varying from 0.1 g/L to 0.5 g/L. They found that the LI-BDV increases or decreases depending on the concentration of NPs. For instance, it is enhanced by 8.2% and 5.3% for concentrations of .1 g/L and 0.2 g/L C<sub>60</sub> with respect to pure natural ester, respectively. While it decreases by 11.1% with a concentration of 0.4 g/L of C<sub>60</sub> NPs.

Siddique et al. [123] investigated the impact of graphene oxide NPs at different concentrations on the breakdown

voltage of soyabean-extracted natural ester oil. They observed adding NPs results in an increase of the AC-BDV of 15% with respect to basic oil. According to these authors, this increase can be due to the hydrophilic nature of graphene oxide leading to the absorption of moisture from the oil by nanoparticles.

Farad et al [127] observed a maximum increase of 5.4% of the LI-BDV when adding Hexagonal Boron Nitride (h-BN) NPs (0.1 wt%) to cottonseed oil (CSO) natural ester with TBHQ antioxidant. The impulse breakdown strength was measured according to the ASTM D3300 standard. An increase of 7.5% Fe<sub>3</sub>O<sub>4</sub> (0.2 g/L), 16.8% with Al<sub>2</sub>O<sub>3</sub> (0.05 g/L), and 13% with SiO<sub>2</sub> (0.2 g/L) towards LI-BDV has been observed. This increase was attributed to this variation to the hydrophobicity of h-BN.

## V. DISCUSSION

It appears from the above that the improvement or degradation of vegetable oils when adding nanoparticles depends on the type of nanoparticle material (conducting, semiconducting, and insulating), their size and concentration, the type of vegetable oil, and the voltage waveform. Various mechanisms and hypotheses (assertions) have been put forward to explain the impact of NPs on the breakdown voltage value of insulating liquids. Most of these mechanisms concern the case where the introduction of NPs improves this value while there is no explanation /interpretation for its degradation.

The process of trapping and de-trapping electrons by nanoparticles is at the center of these mechanisms. Depending on the conductivity/permittivity of NPs with respect to those of hosting liquid and their hydrophobicity, NPs accumulate free electrons at their surface or release them influencing thus the propagation of discharges (streamers) and thence the breakdown.

Different possible processes that could be responsible for the increase in breakdown voltage have been discussed by Usama and Beroual [112]:

(1) The NPs would act as electron scavengers both in the initiation phase and in the propagation phase of the streamers, which would make it possible to raise the initiation threshold voltage of streamers and would slow down their development and consequently would raise the breakdown voltage of the nanofluid. Such a process is impossible because electron scavenger compounds are usually halogenated compounds and it was clearly established that such products (halogens) raise the initiation threshold voltage of streamers increasing the breakdown voltage. However, once generated, these products (halogens) accelerate the streamers and thence reduce the breakdown voltage [137];

(2) The NPs act as an insulating barrier by blocking the charges injected by the electrodes into the nanofluid, thus limiting the mobility of charge carriers toward the opposite electrode. The initiation threshold voltage of the streamers will thus be higher, resulting in an increase in the breakdown voltage;

(3) The NP/host liquid interfaces constitute electric double layers (EDL) which trap the electrons moving toward the opposite electrode and transform them into negatively charged NPS. These charged NPs becoming relatively heavy will move less rapidly than the free electrons. In consequence, they reduce the free electrons amount building a space charge that will decrease the electric field within the liquid and thence weaken the local electric field at the head of streamer channels and slowdown the development of these latter leading thus to the improvement in the dielectric strength (breakdown voltage) of insulating fluid.

Electrons will accumulate at these electric double layers (EDL) until they are saturated. As long as the saturation of this double layer is not reached, electrons continue to accumulate; and the breakdown voltage continues to increase until reaching its optimal value which corresponds to an optimal concentration of NPs. The quantity of accumulated charges depends on the number of nanoparticles and their type (conductivity and permittivity). For a given concentration of NPs, the number of interfaces (double layer) will be higher as the NPs are smaller. After saturation of EDL, the excess electrons will move toward the opposite electrode without being captured; the streamers will be accelerated leading thus to a drop of the breakdown voltage. The processes developed in this third item would be the most likely.

To explain the enhancement of breakdown voltage in the presence of certain types of NPs, several authors [43], [45], [46], [47], [102], [113], [114], [115], [116], [117], [118] advanced the trapping/de-trapping electron process. This latter is linked to the depth of potential wells at the NPs level which depends on the physicochemical properties of these NPs and their geometry (shape and size). By reducing the space charge, this process delays the development of streamers what results in the reduction of their propagation velocity and thence in an increase of the breakdown voltage.

Generally, NP dielectric constant is higher than that of hosting insulating liquid. According to Hwang et al. [116], NPs whose conductivity or permittivity are different from those of the base oil improve the dielectric strength; while adding NPs with a conductivity and permittivity of the same order as those of the base oil leads to the reduction of the breakdown voltage. NPs whose conductivity or permittivity mismatch those of base liquid increase the saturation charges on their interface, which slows down the evolution of the streamers, thence enhancing the dielectric strength of NFs. This mechanism is disputable because beyond a certain concentration of NPs, the breakdown voltage decreases.

## VI. CONCLUSION

Analysis of the various works reported in the literature shows that nanoparticles can improve or degrade the dielectric strength of vegetable oils; the results of measuring the breakdown voltage of nanofluids can be, in certain cases, contradictory. It depends on the type of oil and the physicochemical and geometric properties of the nanoparticles. Only the results with TiO<sub>2</sub> nanoparticles (semiconductor material)

seem to be unanimous and this for all vegetable oils regardless of their origin (i.e. the seeds/plants from which they come); they improve the dielectric strength of insulating oils whatever the concentration of the nanoparticles. A general trend was observed concerning this parameter: the breakdown voltage increases with the concentration of the nanoparticles up to its maximum value and then decreases beyond this concentration. It would be risky to generalize the results obtained for a certain type of nanoparticle or natural ester. It is therefore appropriate to consider, depending on the type of oil, the type of nanoparticles, and the concentration that would give the most efficient nanofluid. These precautions as well as the reserves mentioned above explain why natural ester-based nanofluids are not yet commercialized for transformer oil alternatives.

## ACKNOWLEDGMENT

The authors are grateful for the support by the Researchers Supporting Project number (RSP2024R258), King Saud University, Riyadh, Saudi Arabia.

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