

DOI 10.24425/aee.2024.152115

Key factors enhancing the electrical properties of nanofluids. A mini-review of the applications in the energy-related sectors

OLEKSANDR BOIKO¹✉, HENRYKA DANUTA STRYCZEWSKA¹,
GRZEGORZ KAROL KOMARZYNIEC¹, KENJI EBIHARA² SHIN-ICHI AOQUI³,
MASAAKI YAMAZATO⁴, MYKHAYLO ZAGIRNYAK⁵

¹Lublin University of Technology

Department of Electrical Engineering and Superconductivity Technologies
38A Nadbystrzycka St., 20-618 Lublin, Poland

²Environment and Energy Laboratory

25-39 Suizenji-Park, 862-0956 Kumamoto, Japan

³Sojo University, Faculty of Computer and Information Sciences

4-22-1 Ikeda, 860-0082 Kumamoto, Japan

⁴University of the Ryukyus, Department of Electrical and Electronics Engineering

1 Senbaru, Nishihara, 903-0213 Okinawa, Japan

⁵Kremenchuk Mykhailo Ostrohradskyi National University

Department of Systems of Automatic Control and Electric Drives

20 Pershotravneva St., 39600 Kremenchuk, Ukraine

e-mail: {✉ o.boiko/h.stryczewska/g.komarzyniec}@pollub.pl, ebihara@environment-lab.com,
aoqui@cis.sojo-u.ac.jp, yamazato@eee.u-ryukyu.ac.jp, mzagirn@gmail.com

(Received: 06.08.2024, revised: 24.11.2024)

Abstract: The article presents a mini-review of key factors significantly affecting the electrical properties of nanofluids. One-step and two-step approaches, together with examples of vacuum sputtering-based techniques, chemical reduction, and mechanical mixing techniques, were explained. The crucial factors enhancing the electric and dielectric responses, such as nanofiller concentration, its type, geometry, uniformity of distribution in the base liquid as well as the base liquid's type, temperature, chemical stability, etc., were analyzed. Special attention was paid to the impact of the parameters on electrical conductivity, permittivity, and dielectric losses. The selected models for nanofluid's conductivity prediction have been presented. The potential and implemented applications of nanofluids in the energy-related industry branches with reference to their electrical properties have been reviewed. Examples



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of applications in power transformers, solar cell production processes, nanoelectrofuel flow batteries, and other electrotechnologies have been analyzed.

Key words: dielectric properties, electrical conductivity, energy-related applications, functional materials, nanofluids, nanoparticles

1. Introduction

Currently, the development of electrical materials science, triggered by the issue of sustainable development of the energy sector consisting of the issues of efficient generation, processing, and consumption of electrical energy, as well as its transmission and storage, is based on the search for new unconventional functional materials designed for specific energy applications. A very important group of modern electrotechnical materials studied as a priority are structures on the nanoscale with conductive, dielectric, semiconductive, magnetic, superconductive, and electro-optical (optical fibers) characteristics, the electromagnetic properties of which are much better than those of their macro-scale counterparts [1–6].

Among such a wide spectrum of modern functional nanostructures, liquid nanomaterials deserve special attention due to their physicochemical properties, especially thermal, chemical, and optical ones. Nanofluids (NFs) are considered colloidal suspensions containing nanoparticles (NPs) in a liquid matrix, which is a base fluid (BF). NFs usually contain nanoparticles of pure metals (i.e., Ag, Au, Ni) [1–7] and metal oxides (i.e., Fe_3O_4 , CuO, Al_2O_3) [2, 8–10], silicon oxide [3], carbides (SiC, WC, TiC) [4], and carbon nanotubes and graphene [5, 11–13] of various configurations as nanofillers.

In many cases, water [14–18], ethylene glycol (EG) [2, 15, 17, 19], oils (mineral, natural-type) [4, 10, 20–29], are used as the BFs. The vast majority of the nanofluids studies focus on analyzing and comprehending the behavior of NFs models as heating or cooling mechanisms in various engineering fields, with an accent on the first [14, 30, 31]. It is strictly connected to the thermophysical properties of the NFs, such as thermal conductivity, viscosity, density, heat capacity, etc., that rapidly improve after the introduction of the nanofiller to the BF. The most important key parameters of NFs that cause a significant change in thermal properties include: nanofiller size and shape [23, 32–37], their type and alignment [38], the fact of using surfactants [22, 26, 39], solvent and hydrogen bonding issues [40, 41], NF's temperature [9, 10, 17, 20], the type of BF [38], and the pH level of BF [15, 42–44].

The NFs are also considered long-term promising candidates for applications in the environment and green energy sectors [40]. The use of certain combinations of BF and nanofiller allows one to produce non-toxic, biodegradable, pure, and safe with lower energy consumption NFs via green methods where the nanofiller is produced from the natural extracts.

The most commonly used extracts are: tea leaves, fruits, and vegetables – for green production of graphene [5], CoFe_2O_4 -rGO [32], SiO_2 [16], etc.; flowers and leaves – for obtaining CuO [33], Ag and Se [34], etc.; wood waste – for green production of Tin [41]. A few exemplary applications of green fluids in the energy sectors are listed below:

- replacing the mineral oil with ester oil (characterized by better biodegradability and improved fire points) as a BF in the graphene-contained NFs for power transformer applications significantly decreases transformer aging [12];
- the use of nitrogen doped activated carbon/graphene, produced from the empty fruit bunches pulp, as a nanofiller and EG as a BF, enhances the electrical conductivity by even 11 433% with respect to pure EG [13];
- the Al_2O_3 -contained and cotton-seed oil-based NFs also demonstrate a significant improvement of the dielectric response in comparison to the BF [22].

During the last decade, more and more research has been conducted on the electrical or dielectric properties of NFs. In contrast to thermal issues, research on aspects related to: the impact of electric and magnetic fields on NF on a nano- to macro-scale; the dependence of the electrical response of NFs on the structure and chemical composition; as well as the parameters of both the nanofiller (mono or hybrid type) and the base liquid; and the influence of the experimental conditions (temperature, pressure, atmosphere, etc.) on the electrical parameters of NFs is only gaining momentum. Moreover, in-depth knowledge of the electrical properties of nanofluids is extremely important from the point of view of their engineering applications in the fields of sustainable electroenergetic development, energy-efficient electronics, renewable energy sources, the environment, and bio-nanotechnology.

The main object of the article is to present the newest critically important information on the electrical properties of the NFs, where the main attention will be paid to the key factors having a crucial impact on the DC and AC electrical parameters of the NFs, such as electrical conductivity, resistivity, ACBV, dielectric losses, and permittivity. Section 2 of the article presents a brief discussion of the techniques and approaches (one-step and two-step) for NF preparation. Section 3 is fully dedicated to the description of the impact of the volume concentration, sizes and structure (core-shell), temperature, and sonication parameters such as time, power, amplitude, and frequency, on the NF's electrical characteristics. The relationships between the electrical properties of NFs and their potential and current applications (i.e., transformer isolation, solar cell production, batteries for energy storage, hydrogen production, coolants for micro and microelectronic systems, and others) are presented in Section 4. Section 5 includes suggestions for the future research of electrical behavior of NFs with the main focus on electrical interaction between single NPs, development of the production methods, solving the problem of stability and agglomerations in NFs, modernization of actual mathematical models describing the electrical conductivity and developing new ones taking into account all the drawbacks of the previous ones, optimization of ultrasonication parameters, in-depth study of hybrid, bio-based, and less harmful NFs, developing and upgrading the green methods for NF preparation.

2. Manufacturing methods

The preparation of NFs is an extremely important stage in the experimental studies. Since the NFs are produced by dispersing solid nanofiller particles into a BF, NP agglomerations (cluster agglomerations, islands, grains out of nanoscale) are the major problem [45]. Another important

factor of the NFs during the synthesis is sedimentation, chemical stability and purity of the BF, stability of the synthesis parameters, selection of suitable activators and dispersants, homogeneity and hydrodynamic stability of the suspension, etc. [43].

The vast majority of the papers on NFs present two approaches for their preparation: the one-step method and the two-step method.

2.1. One-step method

The main difference between these methods relies on the combination of issues of nanofiller's production and its dispersion into the BF. During the one-step method (1S), these two stages run simultaneously, as the method's name suggests, into a single step. There are a few deposition techniques, such as laser ablation and magnetron sputtering [46, 47], as well as chemical reduction [48, 49], etc., that allow one to obtain homogeneous NFs with uniform nanofiller.

Choukourov *et al.* [46] used a DC magnetron-based gas aggregation cluster source (GACS) for the synthesis of copper NPs by sputtering a copper target and subsequent deposition of the NPs onto the surface of polyethylene glycol (PEG) liquid (vacuum compatible with an average molar mass of 400 g/mol). The spherical copper NPs with an average size of 20 nm were produced under a plasma current of 150 mA and a deposition time of 15 min, which caused the net concentration of Cu NPs to be 50 µg/ml.

Seifikar *et al.* [47] reported that the preparation of NFs contained Cu, Au, Ag, Ni, Pd, and Pt NPs dispersed in PEG (with an average molar mass of 200 g/mol) via laser ablation in liquid (LAL) technique. LAL is a well-known impurity-free, eco-friendly technique that allows for the production of NPs with controllable size and morphology.

As for the chemical reduction method, Shenoy *et al.* [48] prepared the copper NFs by reducing copper nitrate with glucose in the presence of sodium lauryl sulfate. As a BF, the mixture of water and EG was chosen. The spherical particle fraction in the samples varied from 0.01 wt.% to 0.15 wt.%, while the average size ranged from 19 nm to 58 nm. Zhou *et al.* [49] reported the preparation of Cu/VP-1 NFs, where VP-1 is a heat transfer oil. The initially prepared NaCl and CuSO₄ solutions were mixed and subdued to continuous stirring until a saturated solution of Na₂CuCl₄ formed. Afterwards, it was mixed with a phase transfer agent, which was tetra-n-octylammonium bromide. The reduction reaction was carried out with the use of a sodium borohydride NaBH₄ reducing agent. In order to examine the sedimentation and optical properties of the obtained NFs, Zhou *et al.* used different surfactants such as Span-80, Tween-40, oleic acid, and dodecanethiol. The average particle size of Cu NPs was about 10.6 nm, with a size distribution in the range of 4 nm to 21 nm. The basics of the chemical reduction method are explained in detail in [49].

2.2. Two-step method

In the two-step (2S) method, the nanofiller is obtained by physical or chemical methods as a first step. The second step is dispersing the nanofiller into the BF until the desired NF is ready [50]. A 2S approach is the most frequently chosen method for NFs containing metal oxide nanoparticles or carbon nanotubes [43]. This method is cost-effective and allows for large-scale

preparation of NFs. The major disadvantage of this method is the presence of a noticeable amount of nanofiller's agglomerations in the end product [50].

Akilu *et. al.* [19] prepared EG-based hybrid NFs of TiO₂-CuO/C with different particle concentrations ranging from 0.5 vol.% to 2.0 vol.% and average particle sizes of 26 nm (TiO₂) and 20 nm (CuO/C). TiO₂-CuO/C nanocomposite has been synthesized with the wet mixing method using 80% TiO₂ and 20% copper oxide, as well as 20 mL of hexane for each 1 g of the sample's weight. On the next step, the TiO₂-CuO/C mixture was dispersed in EG.

Sharma *et. al.* [51] prepared double-distilled water-based carbon nanotubes-contained (CNTs) NFs with concentrations of CNTs of 0.274, 0.548, and 0.822 wt.%. The CNTs (multiwalled, with an outside diameter of 50–80 nm and an inside diameter of 5 nm–15 nm) were supplied by The Nanostructured and Amorphous Materials Inc. (USA). To stabilize the CNTs in water, it was necessary to add the surfactant sodium dodecylbenzene sulfonate purchased from HiMedia Laboratories Pvt. Ltd. (India). The mixture of CNTs and distilled water was then ultrasonicated for an hour and additionally centrifuged to reach the desired stability of the suspension. More details and examples of the application of the 2S method are presented in the reviews on the preparation and study of the NFs physico-chemical properties [42, 43].

3. Key parameters enhancing the electrical response

Based on the available literature and knowledge about the latest experiments describing the electrical properties of nanofluids, it can be stated that they depend, for the most part, on the same factors as the thermal properties mentioned in Subsection 1.2. Much greater, even crucial, influence on electrical properties is exerted by the type of nanofiller (or types, if it is a hybrid NF), the type of BF, the volumetric content of the filler and its uniformity of distribution in the BF (this is directly related to the charge transfer mechanisms), the size of particles and their structure (e.g., core-shell), and temperature [2, 3, 8, 9, 17, 23–28, 35]. The importance of the types of NPs and BFs is strictly related to the resistivities of each phase, which results in the total resistivity of NF. The form of particles has a slightly smaller influence on electrical properties, but this can be applied to metals, metal oxides, and carbides. In the case of CNTs, their length is very important from the electrical point of view, which is usually much larger than the diameter and can exceed the nanoscale (100 nm), as reported in [51].

Nanofluids can also be considered as dynamic systems for electrical conductivity. The latter is disadvantaged by the agglomerations as well as the Brownian motion of NPs and their sedimentation, which are also related to the density, pH, and viscosity of the base fluid, surfactants, and other used chemical reagents [42, 43]. Without a good stabilization of the NF's structure, the gradient of cross-sectional electrical resistance becomes more sufficient.

The majority of research papers on NFs report choosing BFs with a dielectric nature and conductivities much higher than the conductivity of nanofiller [2, 4, 8, 14, 15, 21, 30, 31, 39, 45]. The arrangement of more conductive phase particles (pure metals, CNTs, semiconductors, and ferromagnetic particles) inside the isolation phase (BF) resembles very much the metal-dielectric nanocomposites (MDNCs) reported in our earlier studies [52, 53]. With one basic difference:

MDNCs have a stable static bulk structure, and they also contain the ferromagnetic alloy (FeCoZr) or conductive copper NPs in dielectric matrices from PbZrTiO₃ (also known as PZT), CaF₂, Al₂O₃, and SiO₂. This kind of structure under an external electric field can demonstrate special charge carrier mechanisms such as hopping conduction and tunneling, as well as percolation, resonant, and non-coil inductance properties [52]. It also possesses interfacial and dipolar polarization mechanisms and near-Debye relaxation [53]. All of the above-mentioned aspects have not yet been thoroughly studied in the NFs, which indicates an interesting gap in the complete understanding of NF properties.

3.1. Impact of key parameters on the electrical conductivity

Fal *et al.* reported the preparation of EG-based TiO₂ NFs via the 2S method, as described in [2]. Three types of TiO₂ powder with anatase phase (A), rutile phase (M1), and a mixture of both phases (M2) were used during the preparation. The TiO₂ average particle sizes in accordance with the types were 4 nm–8 nm, 2 nm, and 21 nm, respectively. The obtained samples demonstrated a volume fraction concentration in the range of 1 wt.%–7 wt.%. The authors point to a crucial impact of the nanofiller and temperature on the electrical conductivity of the EG-TiO₂ NF, as shown in Fig. 1(a). Figure 1(b) presents fitting results obtained on the basis of existing theoretical models and approaches. As can be seen from Fig. 1(a), an increase in volume fraction (φ_v) causes a further significant increase in conductivity, showing a maximal enhancement at 45°C of about 1300% (black labels). An increase in temperature from 10°C to 45°C results in an increase in conductivity of about 300%.

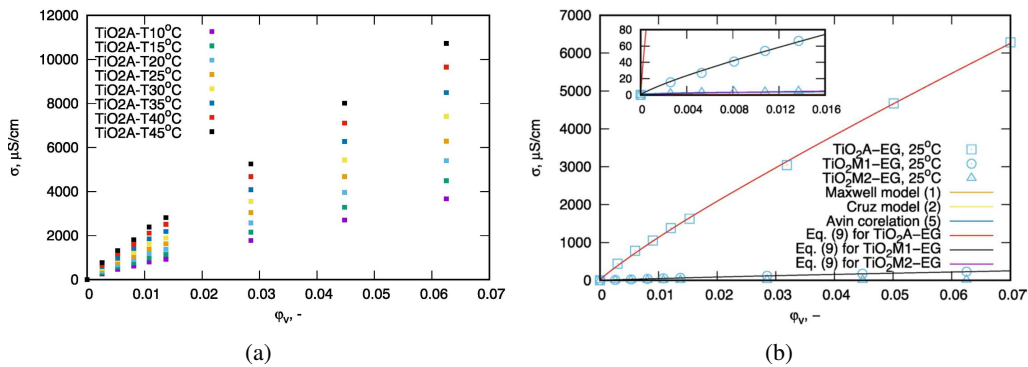


Fig. 1. Electrical conductivity of EG-TiO₂ NFs [2]: (a) impact of volume fraction φ_v and temperature for type A NF; (b) comparison of obtained results to the Maxwell, Cruz, and Avins theoretical models. Solid lines demonstrate fitting in accordance with the used equations

The solid lines express fitting calculations using the following models and theoretical investigations on the electrical conductivity of NFs:

- Maxwell model [54] (orange line):

$$\sigma_{nf} = \left(1 + \frac{3(\alpha - 1)\varphi_v}{((\alpha + 2) - (\alpha - 1)\varphi_v)} \right) \sigma_{bf}, \quad (1)$$

where: $\alpha = \sigma_p / \sigma_{bf}$, σ_{nf} is the conductivity of NF, σ_p and σ_{bf} are conductivities of particles and BF, respectively. This model has a few drawbacks: it doesn't take into account NPs motion, size, and surface area, which can affect conductivity. Moreover, it assumes only the spherical-form particles and their random arrangement, with the distances much larger than their average size;

- Cruz *et al.* model [8] (yellow line, calculated in accordance with case (c)) that includes three situations:

a) nanofiller's conductivity is much lower than the conductivity of BF

$$\sigma_{nf} = \left(1 - \frac{3}{2}\varphi_v\right) \sigma_{bf}, \quad (2)$$

b) σ_p and σ_{bf} are comparable

$$\sigma_{nf} = \sigma_{bf}, \quad (3)$$

c) nanofiller's conductivity is much higher than the conductivity of BF

$$\sigma_{nf} = (1 + 3\varphi_v) \sigma_{bf}, \quad (4)$$

- Awin *et al.* model (blue line) proposed in [55] where φ_v and temperature T ranges of about 0.0015%–0.005% and 25°C–45°C were taken into account

$$\sigma_{nf} = 0.459T^{0.968}\varphi_v^{0.24}. \quad (5)$$

This model also doesn't consider the particle size impact on the conductivity; it was verified on the Al_2O_3 , TiO_2 , and CuO NPs dispersed in the ultra-pure water. The diameter of spherical NPs varied from 10 nm to 20 nm;

Islam *et al.* model [17] (other lines):

$$\sigma_{nf} = (11.214 + 2.626 \ln \varphi_v + 0.2371 \ln T) \sigma_{bf}. \quad (6)$$

The Eq. (6) was as well verified in the case of NFs based on a 50/50 mixture of deionized water and EG containing TiO_2 NPs with an average size of 40 nm. In the study, the maximum attention was paid to temperature impact on the conductivity.

Fal *et al.* [2] used all the above-mentioned models to analyze the obtained results on their own EG- TiO_2 NFs and stated a significant discrepancy between measured and theoretical data. The authors proposed the original power function for conductivity based on the mass fraction φ_m and numerical factors A and B :

$$\sigma_{nf} = \left(1 + B\varphi_m^A\right) \sigma_{bf}. \quad (7)$$

The well-known fact is that the conductivity of NFs depends significantly on temperature T , as shown in Eqs. (5) and (6). Despite this fact, the authors were able to obtain Eq. (7) with no T parameter. The proposed power function gave very good accuracy in comparison results for all types of NFs.

Similar situations with a significant increase in conductivity after adding a better-conducting nanofiller to BF were also reported in the works of [23, 35, 42, 43]. Du *et al.* [23] describe how the size of Fe_3O_4 NPs affects the volume resistivity (that is opposite to conductivity) of vegetable

oil-based Fe_3O_4 NFs. The authors report a not-so crucial but noticeable ($\leq 70\%$, especially for frequencies less than 0.1 Hz) increase in resistivity while the NP size changes from 8.6 nm via 15.2 nm to 24.4 nm. This also means that the increase in size results in conductivity decay. Krishnan *et al.* [35] demonstrate how particle size and temperature affect the distilled water-based $\gamma\text{-Al}_2\text{O}_3/\text{MWCNT}$ NF's conductivity. The average sizes of 5 nm for MWCNT and 5 nm and 30 nm for Al_2O_3 were chosen for the experiment. The authors noticed that 30 nm alumina-based NFs result in lower conductivity than 5 nm alumina NFs. The reason could be the change in effective particle charge density as the size gets smaller.

Yasmin *et al.* [44] and Giwa *et al.* [18] describe on the example of hybrid MgO-ZnO/DIW NFs with the nanofiller's concentration of 0.5 vol.% the effect of sonication time, sonication amplitude, and pH on the electrical conductivity. Based on [18, 44], it can be concluded that the conductivity rises when sonication time is extended from 30 min to 45 min. After exceeding 45 min of sonication, the conductivity starts to decay while the sonication time increases. In the case of pH impact, the lowest pH corresponds to the highest conductivity. After the sonication time increases from 45 min to 120 min, the pH increases rapidly from approx. 10.72 to 10.86. The tested sonication amplitudes range of 70%–80%, and the sonication effects on electrical conductivity and pH allow one to define the optimized parameters for the improvement of 0.5 vol.% NF's stability at the CMC point. Those parameters are: 45 min of sonication time, 10.72 pH, and 75% of sonication amplitude.

Hewitt *et al.* [36] reported the effects of high-energy probe sonication (HEPS) on the electrical conductivity of multiwalled carbon nanotubes (MWCNTs)-deionized water NFs in the temperature range of 20 K–290 K. The MWCNTs with an average diameter of 150 nm were synthesized by chemical vapor deposition at 850°C. The 25 mg of raw MWCNTs were dispersed in 15 mL of water, which gives a nanofiller's concentration of 1.67 mg/mL. HEPS time of 15 min to 12 h was used to compare the electrical conductivity of raw and sonicated NFs. The experiment showed that the highest value of conductivity was recorded after the 15 min of sonication, while from approx. 28.5% to approx. 41.7% lower was the conductivity of raw fluid. Further extension of HEPS resulted in noticeably lower values (than the row one's) of conductivity with a decreasing tendency while the sonication time increased from 1 h to 12 h via 3 h and 6 h. Moreover, the temperature increase caused a significant increase in conductivity, which was enhanced with a decrease in sonication time. It is also worth noticing that the HEPS applications drastically lower the length of MWCNTs, especially longer ones (i.e., 18 h), from at least 60 μm to even 1–5 μm , which can cause a noticeable impact on the electrical conductivity.

The vast majority of papers on the electrical conductivity of NFs report an increasing tendency with the volume fraction [18, 23, 24, 35, 36, 41–44]. However, if the electrolyte is added to the NF, the opposite dependence can be observed. Sarojini *et al.* [56] produced alumina-water NFs with nanofiller concentrations ranging from 0.05 vol.% to 1.6 vol.%, and then mixed them with hydrochloric acid (HCl) with three different concentrations of 1 M, 0.5 M, and 0.05 M. The experiment showed that in all cases the conductivity decreases (the highest reduction of about 3 times was noticed in the case of 1 M) until reaching the critical concentration, when the volume fraction increases. For the concentration higher than critical, the slight increase in conductivity is registered. Moreover, an increase in electrolyte concentration refers to the higher critical concentration value. The paper [56] also demonstrates, that there is no temperature dependence of the conductivity for the alumina-water NFs with concentrations < 0.5 vol.%. For the higher concentrations, temperature dependence of conductivity became significant.

Cieśliński *et al.* [10] also reported percolative temperature dependence of electrical conductivity of thermal oil-based Al_2O_3 NFs as well as decreasing temperature dependence of conductivity of TiO_2 NFs with the same BF. In the case of thermal oil (TO) and TO- Al_2O_3 fluids, the conductivity rises until it reaches local maximum at 29°C. Further temperature increases reduce the conductivity up to the starting level. The stable decay in the whole temperature range can be observed in the case of TO- TiO_2 NFs. Furthermore, paper [10] reports a significant decrease in conductivity with the nanofiller's concentration of TO- Al_2O_3 NFs in the temperature range of 20°C–40°C. For this purpose, the NFs with concentrations of 0.1 vol.%, 1.0 vol.%, and 5.0 vol.% have been prepared. The highest drop in the conductivity was registered for 29°C, where the conductivity of TO was measured as $\approx 0.71 \mu\text{S}/\text{cm}$, and the conductivity of 5.0 vol.% TO- Al_2O_3 was $\approx 0.29 \mu\text{S}/\text{cm}$.

3.2. Dielectric properties of nanofluids

3.2.1. AC breakdown voltage

A significant number of articles from the past decade have been dedicated to the investigation of the dielectric strength and stability of oil-based NFs [12,24,25]. Koutras *et al.* [24] investigated AC breakdown voltage (ACBV) in the FR3 transformer oil-based TiO_2 NFs prepared by the 2S approach. The nominal diameter of NPs was 21 nm, and the final volume concentrations of the obtained samples were 0.005 wt.%, 0.010 wt.%, 0.020 wt.%, and 0.040 wt.%. An investigation of ACBV was conducted with the use of a Baur DTA 100C generator, allowing it to produce voltage up to 100 kV with a pulse frequency of 60 Hz. The experiment showed that the ACBV increased with the increase in TiO_2 concentration. The highest value of ACBV was demonstrated by a 0.020 wt.% sample; the improvement in this case was found to be about 22.4% compared to the FR3 value. The ACBV improvement when the NPs concentration rises can be referred to as the increasing in shallow trap density by the volume fractions and NPs agglomerations, which leads to the delay of streamer propagation via the electron trapping path as well as de-trapping processes that reduce the NF's charge mobility.

Hussain *et al.* [25] investigated the dielectric properties of NFs containing three types of nanofiller: iron phosphide, iron (II, III) oxide, and cobalt (II, III) oxide, in BFs of synthetic ester oil and natural ester oil. Samples demonstrated NP concentrations ranging from 0.0015 wt. to 0.004 wt.% and particle sizes of 50 nm–100 nm (iron oxide), < 50 nm (cobalt oxide), and 50 nm–100 nm (iron phosphide). The measurements have been conducted with the use of an oil breakdown tester in accordance with IEC 60156 and two standardized Verband Deutscher Elektrotechniker electrode configurations: mushroom-mushroom (M-M) and sphere-sphere (S-S). Figure 2(a) presents the results of ACBV measurements for iron (II, III) oxide NFs. It can be seen that characteristics for natural ester oil show a continuously increasing trend, with the highest value being reached at the highest nanofiller's concentration, demonstrating a maximum improvement in ACBV of about 33.5% (S-S). For the synthetic oil, the highest value of ACBV is observed in a 0.0025 wt.%, which gives a maximum improvement in ACBV of 30.7% (S-S).

Farade *et al.* [12] used cottonseed oil (CSO) oil-based multilayered graphene sheets (MGS, 5–10 micron lateral dimension, 3 nm–6 nm thickness) to demonstrate the concentration and sonication time impact on the ACBV. The concentrations of the MGS were estimated at 0.0015 wt.%, 0.003 wt.%, 0.006 wt.%, and 0.01 wt.%. Figure 2(b) shows the results that have a similar tendency as in the above-described cases: the ACBV increases while the concentration increases.

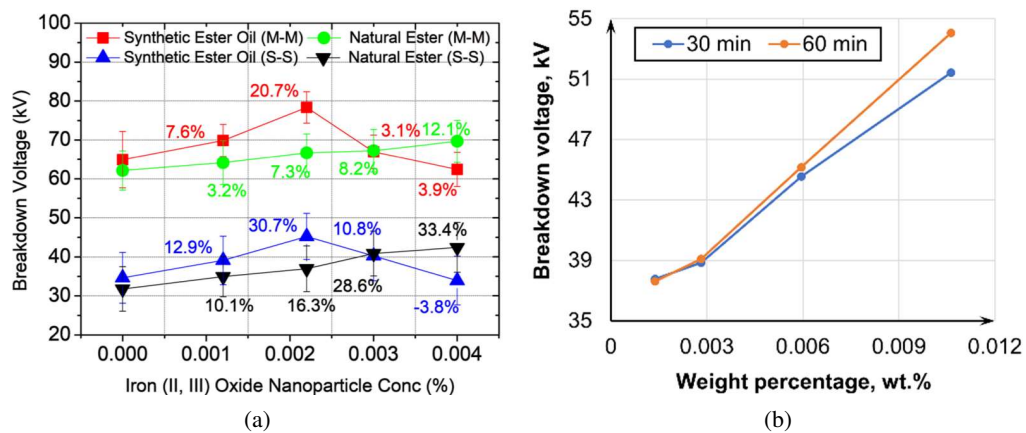


Fig. 2. ACBV measurements for: (a) synthetic and natural ester oil-based iron (II, III) oxide NFs [25]; (b) cottonseed oil-based MGS NFs [12]

Increasing sonication time from 30 min to 60 min increases ACBV. This tendency is intensified when the mass fraction of the nanofiller grows, with the highest improvement of 5.13% for the 0.01 wt.%. Another example of impact of sonication time on ACBV of oil-based CNTs NFs is presented by Farade *et al.* [29]. The authors used sonication time duration of 30 min and 120 min, and CNTs concentrations of 0.01 wt.%, 0.03 wt.% and 0.05 wt.%. The experiment showed the ACBV enhancement while the time increased in all three cases. The maximum improvement of 25% (from 34.56 kV to 43.25 kV) was registered in the 0.03 wt.%.

Table 1 summarizes an impact of volume concentration, weight fraction and sonication parameters on the ACBV of the described in the article oil-based NFs produced by 2S method.

Table 1. Impact of volume fraction and sonication time on ACBV of selected NFs

Ref.	Nanofiller	Concentr.	Av. size	Important findings regarding ACBV and dielectric strength
[24]	TiO ₂	0.005–0.040 wt.%	21 nm	Increase in volume fraction of TiO ₂ causes significant increase in ACBV with a maximum improvement of 22.4% for 0.020 wt.%.
[25]	Fe ₃ O ₄ , FeP, Co ₃ O ₄	0.0015–0.004 wt.%	50–100 nm	Natural ester oil shows a continuously increasing trend in conductivity while the weight fraction increases. The highest improvement is about 33.5% (0.004 wt.% sample) for sphere-to-sphere electrodes arrangement. In the case of mineral oil, the highest ACBV improvement of about 30.7% is registered for the 0.0025 wt.% sample.

Continued on next page

Table 1 – Continued from previous page

Ref.	Nanofiller	Concentr.	Av. size	Important findings regarding ACBV and dielectric strength
[12]	MGS	0.0015–0.01 wt. %	3–6 nm/ 5–10 micr.	ACBV improvement of about 42% at highest concentration of 0.01 wt. %. The sonication time shows a visible impact on ACBV after exciding 0.006 wt. %.
[27]	CCTO*	0.001–0.05 vol. %	30–200 nm	Percolative dependency of ACBV on volume fraction. The critical concentration of CCTO is 0.005 vol. %.
[29]	CNTs	0.01–0.05 wt. %	< 8 nm, > 2 nm	Increase in sonication time from 30 min to 120 min improves significantly the ACBV with a maximum enhancement of 25% for 0.03 wt.
[28]	Fe ₃ O ₄ , TiO ₂ , Al ₂ O ₃	–	10 nm	Introduction of Fe ₃ O ₄ , TiO ₂ , Al ₂ O ₃ NPs with some certain (unprecise) concentration causes an enhancement in ACBV of about 44.3%, 33.3%, 35.5%, respectively.
[37]	Fe ₃ O ₄	–	15 nm, 24 nm, 42 nm	The enhancement of the ACBV of the insulation paper increased by about 10% after adding the nanofiller to FR3 oil.

*CaCu₃Ti₄O₁₂

3.2.2. Electric permittivity and dissipation factor

Farade *et al.* [26] prepared CSO-based Al₂O₃ NFs with the use of SDBS surfactant. The weight percentage of the nanofiller in the obtained samples was as follows: 0.01 wt. %, 0.02 wt. %, 0.03 wt. %, and 0.05 wt. %. The Al₂O₃ particle size varied from 15 nm to 25 nm. Fig. 3(a) describes the relative permittivity dependence on nanofiller concentration. The permittivity increases, with a maximum enhancement of about 30%, while the weight fraction of Al₂O₃ also increases. There is also a visible slight temperature effect on permittivity that improves by about 10% while the temperature rises from 45°C to 90°C. The paper also reports that the increase in sonication time from 30 min to 60 min leads to a slight increase in dielectric constant (especially for lower nanofiller concentrations) by approx. 0.5% on average. A quite similar situation is presented in Hussain *et al.* [25], where synthetic ester oil-based iron phosphide, iron (II, III) oxide, and cobalt (II, III) oxide NFs were tested (Fig. 3(b)). In the case of iron phosphide and cobalt (II, III) oxide NFs, increasing the nanofiller's concentration from $\approx$ 0.0013 wt. % to 0.003 wt. % results in an enhancement of about 6.89% and 3.24%, respectively. In the case of iron (II, III) oxide, the maximum enhancement is registered in a 0.002 wt. % sample. For all three nanofillers, the samples with 0.004 wt. % demonstrate a significant reduction in permittivity, even below the synthetic oil level. This fact can be related to unstable nanoparticles in the solution or the presence of their agglomerations. Similar behavior of NF permittivity vs. nanofiller concentration was also reported in the works [27, 28].

An increase in the Al₂O₃ NPs concentration in CSO oil [26] also leads to a gradual decrease in dielectric losses (Fig. 3(c)). The degradation of dielectric loss tangent ($\tan \delta$) enhances when the temperature is getting higher. It can be explained with respect to the natures of both phases: low-conductivity BF and higher-conductivity nanofiller. The maximum decrease of about 50% is observed in a NF subdued to the highest temperature of 90°C.

Du *et. al.* [3] fabricated FR3 oil-based Fe_3O_4 - SiO_2 hybrid NFs with a core-shell structure of nanofiller. It was reported that the SiO_2 shell had significant effects on the dielectric loss. The shell's thickness varied from ~ 7.5 nm (Fig. 3(d) A) to ~ 50 nm (Fig. 3(d) C) via ~ 24 nm (Fig. 3(d) B). The size of the Fe_3O_4 core was constant, with a value of ~ 60 nm. Fig. 3(d) describes the dependence of the dissipation factor (DF) on the measurement frequency, which ranges from 10^{-2} Hz to 10^7 Hz. The decrease in SiO_2 shell results in an increase in DF that is clearly seen in the frequency range of 10^{-2} Hz to 10^0 Hz. The higher frequencies also demonstrate an increasing tendency (inset in Fig. 3(d)), but this is hardly noticeable without using a log scale.

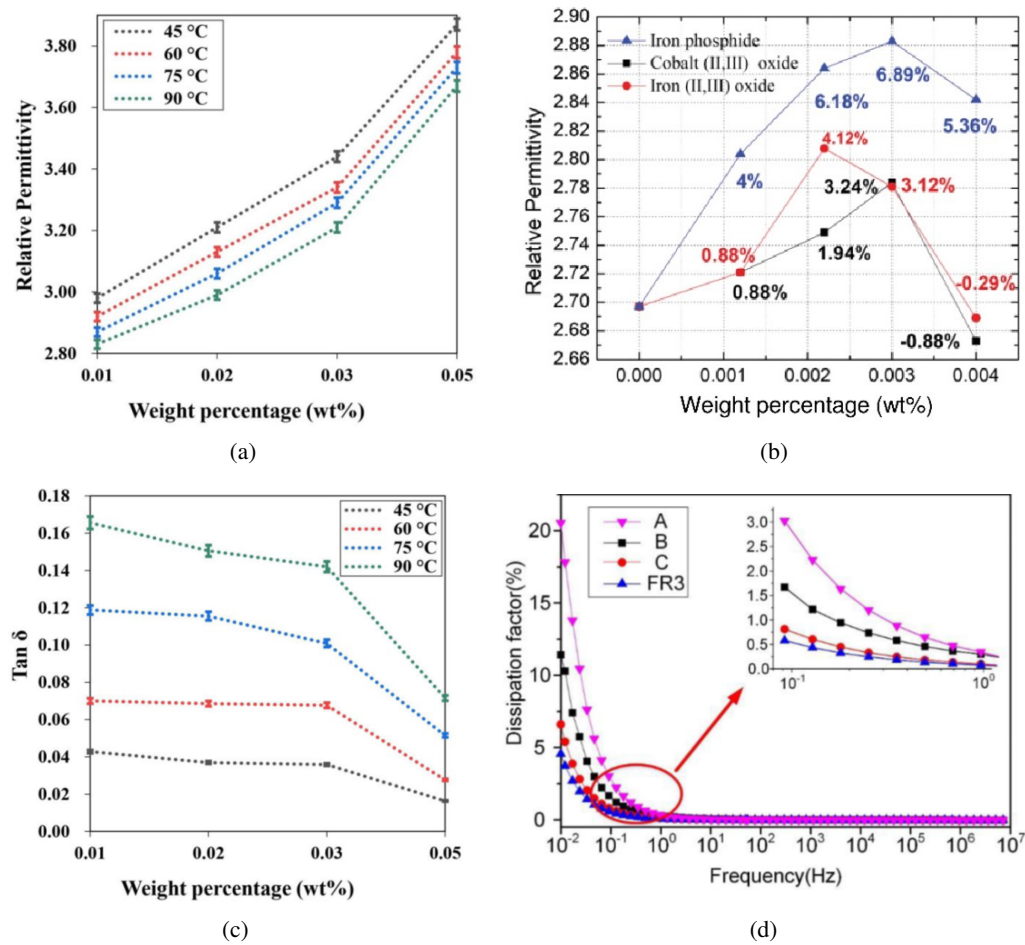


Fig. 3. Dielectric properties of selected NFs: (a) relative permittivity vs. weight percentage of CSO-based Al_2O_3 NFs [26]; (b) relative permittivity vs. concentration of synthetic ester oil-based iron phosphide, iron (II,III) oxide, and cobalt (II,III) oxide NFs [25]; (c) dielectric loss tangent vs. weight percentage for CSO-based Al_2O_3 NFs [26]; (d) dielectric loss vs. frequency for FR3 oil-based Fe_3O_4 - SiO_2 hybrid NFs [3]

Chakraborty *et al.* [9] studied the AC dielectric properties (in the frequency range of 10^{-3} Hz– 10^3 Hz) of TiO_2 and Al_2O_3 NFs based on mineral and FR3 vegetable oils. It was found that the DF decreased after introducing a nanofiller to the oil volume. Moreover, the temperature growth causes a significant enhancement in DF of even about two orders of magnitude (in the case of Al_2O_3 NFs). Al_2O_3 nanofiller weakens more NFs DF than TiO_2 , mainly due to the conductivity differences of both oxides.

Farade *et al.* [29] based on effective literature analysis, he summarized the influence of ultrasonic parameters such as time, power, frequency, amplitude and temperature, on the dielectric properties of oil-based NFs as shown in Fig. 4.

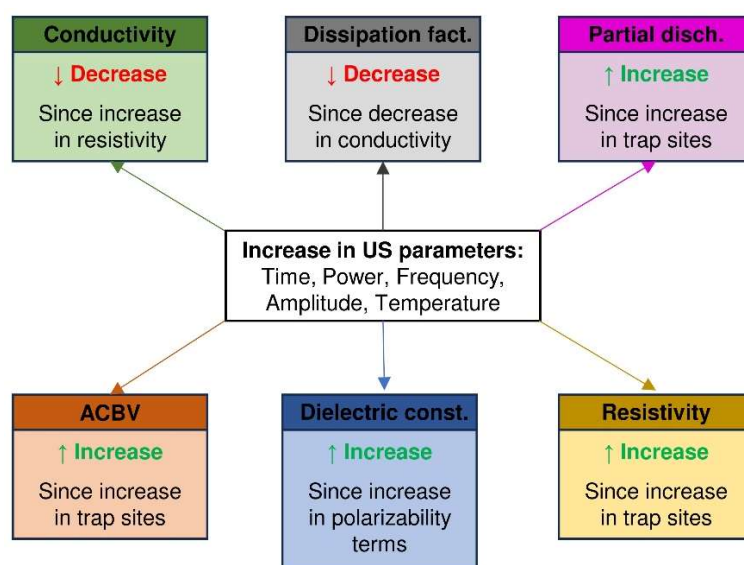


Fig. 4. Impact of the main ultrasonic parameters on the dielectric properties of NFs. Developed based on [29]

An increase in the ultrasonic parameters causes a significant increase in ACBV, dielectric constant, resistivity and partial discharge. This situation can be explained by increased trapping sites for electric charge caused by a higher level of broken agglomerations into smaller particles or clusters and their more even distribution in the BF volume. This also results in the NF's stability enhancement and corresponds to higher zeta potential and slower sedimentation rate. In the case of conductivity and dissipation factors, they are decreasing while the sonication parameters increase. Paper [29] defines these dependencies as directly (ACBV, dielectric constant, resistivity and partial discharge) and inversely (conductivity and dissipation factor) proportional. This is the authors' conclusion, but a number of works [10, 36, 44, 51] report dependencies confirming this statement in terms of their tendency, but disproportionate in terms of the form of the dependency. This is definitely a task that requires additional attention from scientists and further experiments.

Table 2 summarizes an impact of volume concentration, weight fraction, size, and sonication parameters on the dielectric properties of the described in the article oil-based NFs. All included NFs were produced with us of 2S approach.

Table 2. Impact of volume fraction, size and sonication time on permittivity and dissipation factor

Ref.	Nanofiller	Concentr.	Size	Important findings regarding dielectric properties
[26]	Al ₂ O ₃	0.01–0.05 wt. %	15–25 nm	Increase in weight fraction of Al ₂ O ₃ causes an increase in electrical permittivity with a maximum improvement of about 30%. Slight temperature effect on permittivity that improves it by about 10%.
[25]	Fe ₃ O ₄ , FeP, Co ₃ O ₄	0.0015–0.004 wt. %	50–100 nm	In the case of FeP and Co ₃ O ₄ NFs, the growth in concentration results in permittivity improvement of about 6.89% and 3.24%, respectively. All NFs demonstrate percolative permittivity dependencies with a critical value of wt. % of 0.002–0.003.
[27]	CCTO*	0.001–0.05 vol. %	30–200 nm	$\tan\delta$ decreases exponentially (about 3.3 times when 0.001 vol. % and 0.05 vol. % are compared) with increase in concentration of nanofiller. The dielectric permittivity doesn't show clear dependency on volume fraction. The highest value is observed for 0.0025 vol. % and the least value for 0.005 vol. %.
[3]	Fe ₃ O ₄ -SiO ₂	0.1 wt. %	60 nm/ 7.5 nm, 24 nm, 50 nm	Strong frequency dependence of permittivity for the frequencies above 1 Hz. After exceeding this value the permittivity stabilizes. An increase in volume concentration results in the increase in permittivity as well as in dissipation factor.
[9]	TiO ₂ , Al ₂ O ₃	0.2 g/l	20–30 nm	Strong decreasing tendency of DF along with frequencies ranged from 10 ⁻³ –10 ³ Hz. Introduction the nanofiller to BF provides the reduction of DF of about 0.3 order of magnitude for both nanofillers. Increase in temperature improves significantly the DF.
[37]	Fe ₃ O ₄	–	15 nm, 24 nm, 42 nm	Increase in NPs size leads to the decrease in dielectric constant and increase in dielectric losses.

*CaCu₃Ti₄O₁₂

4. Selected applications of nanofluids in electrical engineering

A significant number of reports on the electrical applications of NFs originate from their enhanced dielectric properties. The NFs are widely studied from the point of view of their application in transformer technologies, as shown in [4, 20, 21, 24, 25, 27, 28, 39]. As it was described in Subsection 3.2.a, the introduction of nanofiller increases the ACBV of the transformer oil significantly. Fig. 5(a) describes the process of transformer insulating paper impregnation with FR3 oil-based Fe₃O₄ NFs [37]. Three NFs types with particle sizes of 15 nm, 24 nm, and 42 nm have been used for impregnation. In all three cases, the enhancement of the ACBV of the insulation paper increased by about 10%. It was also noticed that an increase in particle size causes a reduction in the dielectric constant of paper and growth in dielectric losses.

(where PDADMAC is poly(diallyldimethylammonium chloride)) and CuO/PSS (where PSS is poly(sodium 4-styrenesulfonate)) NFs [59]. The NF enhances ion production in the electrolyte during the charging and recharging processes. The advantages of NFB technology can be considered as follows: very good thermal conductivity, power and energy section separation, fast recovery time, and long-life cycle. Nevertheless, it also has some drawbacks: low electrode contact, pressure drop in the cell stack, and turbulent flow problems.

Additionally, the literature contains considerations on the following applications of nanofluids:

- liquids for photovoltaic panel cooling systems [60]. The use of a mixture of water and Al_2O_3 as a cooling agent in traditional polycrystalline solar panels decreases drastically (even about 23%) the panel's operation temperature, which increases their efficiency afterwards;
- hydrogen production through electrolysis [61]. An addition of a small amount of carbon black (≈ 0.1 wt%) to the sodium hydroxide electrolyte causes an improvement in hydrogen production during the electrolysis of about 23.62% due to the enhancement of NF's electrical conductivity;
- liquid magnetic cores for the transformers made from ferromagnetic NFs [62]. Using diesel oil-based Fe_3O_4 NFs instead of bulk magnetic cores increases the inductance, coupling coefficient, and resistance of the transformer's coil as well as decreases its quality factor due to the phase delay between the magnetic field and the magnetization;
- permanent magnetic actuators for microdevices [63]. The water-based Fe_3O_4 NF can be used in rotating permanent magnetic actuator-based micropumping systems in order to enhance their magnetic properties. An increase in volume fraction of Fe_3O_4 NPs from 1 vol.% to 3 vol.% improves the NF's flow rate of about three times, which also increases the viscosity of about 1.4 times;
- effective coolants for electronic devices and systems [7]. The use of de-ionized water-based Ag/ Fe_3O_4 hybrid NFs in thermal management systems of electronic devices as active coolants increases the thermal dissipation efficiency of about 12%, which causes an improvement in cooling efficiency of about 71.5% in comparison to water-cooled systems;
- thermoelectric pipes [64]. The use of electrolyte from deionized water-based TiO_2/NaOH hybrid NF instead of pure water increments significantly its current density (at least for five times for the 25°C) and power density (at least for nine times for the 25°C). The tendency is rising along with temperature.

5. Future research

The complexity of variables defining the electrical behavior of the NF stimulates the ongoing research to move forward and use more precise and qualitative analysis. It would seem that the conductivity of NF is proportionally dependent on the nanofiller, which is usually more conductive than the BF [18,23,35,36,42–44], but there are numerous papers reporting a completely or partially opposite situation [1, 10]. A similar unclear situation can be noticed in the case of dielectric parameters of NFs [18,44] as well as the study of sonication treatment on them [10,29,36,44,51]. There is absolutely no doubt that the type of base liquid and nanofiller, as well as the surfactants and other chemical reagents used to achieve high stability and uniform particle distribution, have

a huge and, in many cases, decisive influence on the electrical properties of NFs [4,38]. Other equally important parameters are temperature, size, and structure, as well as the distribution of the nanofiller in the BF volume [3,20].

Nevertheless, there are some gaps in the picture of a thorough understanding of the electrical nature of NFs, which indicate the need for further in-depth research in the following (but not limited to) directions:

- *electrical interaction at the level of individual particles separated by a BF.* It would be necessary to consider the mechanisms of charge transfer between particles, the mechanisms of dielectric polarization and relaxation depending on the applied electric field, the influence of possible structure (e.g., core-shell) and form of particles (e.g., cuboid, octahedron), counteracting sedimentation;
- *the influence of the production method on the oxidation level of metallic or metallic alloys (it can also have a core-shell structure) nanoparticles.* The oxygen coating is an additional potential barrier to be overcome by the electric charge in order to contribute to conductivity. If specific applications of the produced nanofluids are planned, e.g., in memristive switching devices [65], the lowest possible oxidation level should be sought;
- *obtaining the best possible stability* of the NF and its lifetime (until the final sedimentation and loss of designed electrical properties). This is a very important topic from the point of view of the applications of nanofluids in industry;
- *improvement of mathematical models describing electrical processes and phenomena in NFs.* As shown in Section 3.1 on the example of modeling the conductivity of nanofluids [2, 8, 17, 54, 55], the simulation results only partially match the experimental results. Most existing models do not assume the dependence of electrical conductivity on the nanofiller size, and some are only based on the conductivities of individual phases, nanoparticle content, and numerical coefficients without taking into account the effect of temperature;
- *optimization of ultrasonication parameters.* The works [12, 18, 25, 26, 29, 36, 44] show a significant influence of each of the ultrasonication parameters on the dielectric properties of NFs, which means that their optimization is necessary in order to obtain samples with the assumed characteristics;
- *in-depth study of hybrid, bio-based and less harmful NFs.* Since the hybrid NFs contain more than one type of nanofiller, an interaction between different-type nanoparticles while investigating a physico-chemical properties should be taking into the account. The studies should also consider different BFs and efforts in best possible stability via optimization of surfactant adding, pH control and ultrasonication issues [19, 35]. Very important for both hybrid and bio-based NFs are effective production techniques and precise composition investigations [43];
- *green methods for NF preparation.* Green nanofluids are produced using techniques and methods that are clean, environmentally friendly, safe, energy-saving, cost-effective, and have the minimum possible level of danger to living organisms [66]. They should be subjected to more precise and frequent testing than their novel counterparts, if only because of their green applications.

6. Conclusions

Nanofluids are very important from the scientific and engineering applicability point of view of functional nanomaterials. Despite the fact that the thermal properties of nanofluids have been almost thoroughly understood, the electrical properties, processes, and phenomena definitely require the continuation of research to contribute to the development of modern intelligent electrotechnologies and electrical materials science in the field of liquid materials.

The methods of NF's production are well designed and are based on many years of experimental efforts by researchers, but they still need to be improved in order to obtain chemical stability, purity, homogeneity, and hydrodynamic stability of the NFs, as well as to avoid the aggregating of the nanofiller, Brownian motion, and sedimentation.

The 2S method is definitely more often used for the production of NFs. This is due to the lower technological advancement of individual processes and also greater control over them, cost efficiency, large-scale producing, wide commercial availability of NF components (base liquids, nanofillers, surfactants). The 2S method allows for obtaining homogeneous and more stable NFs. Based on metal oxides and CNTs as well as natural and synthetic oils, it is most often used for the production of NFs containing metal oxides or CNTs as a nanofiller, with strictly dielectric properties or in order to enhance them.

The 1S method is more often chosen to avoid oxidation of the nanofiller during production, especially in the case of metals or metal alloys. This is a technique intended rather for laboratory (fundamental) research than for industrial mass production. It requires the use of specialist equipment, but in return it allows easy manipulation of the dimensions and geometry of the nanofiller, which is not possible in the case of commercial purchase. This means that it is a good candidate for the verification of mathematical models describing the mechanisms of electrical conductivity, polarization and dielectric relaxation of NFs. Compared to the 1S method, the disadvantage is the large presence of agglomeration and maintaining stability, which has a significant impact on electrical properties.

The key parameters demonstrating a significant impact on the electrical parameters of NFs are determined as types of BF and nanofiller, nanofiller's concentration, structure, size, and form, homogeneity, distribution of nanofiller in the BF, temperature, sonication treatment, surfactants, and other chemical precursors used during the preparation. The introduction of more conductive nanofiller, even in small amounts (less than 0.1 wt.%), to the less conductive (rather insulative) BF immediately causes a significant increase in the NF's conductivity. A similar increasing tendency repeats in the case of electric permittivity, which increases as the nanofiller concentration increases. The dissipation factor demonstrates the opposite dependency. In the vast majority of cases, the growth of nanofiller's concentration results in an increase in the breakdown voltage of the NFs, which is a very important aspect for the potential applications of NFs. Fig. 6 summarizes the influence level of key parameters, where the most influencing are on top and the least influencing (but still significant) are on the bottom. With no doubt, the types of NF's ingredients and nanofiller's concentration are the most valuable. Nanofillers geometry and structure demonstrate much smaller impact, but they cannot be completely ignored during the detailed analysis of the electrical properties of NFs.

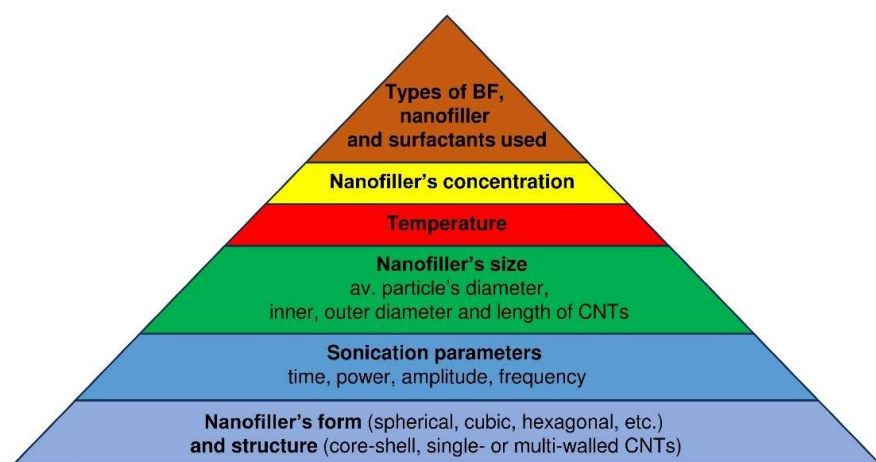


Fig. 6. Graphical representation of hierarchy of the influence of the key parameters on the electric properties of NFs. The impact grows from bottom to top

Taking into account the available knowledge of the electrical properties of NFs, a lot of potential and actual applications have been proposed. Due to the ability of NFs to significantly increase the ACBV of transformer oil by introducing metal oxide NPs or CNTs into it or insulating paper impregnation by the NFs, they can be used in power transformers or magnetic coils, where the NFs replace a regular bulk ferromagnetic core. An increase in ACBV and providing an acceptable stability of the NF can prolong sufficiently the transformer's exploitation time.

Researchers propose applying NFs with semiconducting nanofillers during solar cell production to enhance their efficiency. If the NFs are used as coolants for cooling systems of PV panels, they drop significantly the PV panel's working temperature, which results in better green energy generation and their efficiency in total. When the NFs are used during the solar cell production processes as an interlayer, it enhances the PV effect of the cell, which also improves its efficiency.

An improvement in electrical conductivity, current, and power densities after using the NFs as electrolytes for electrolysis processes allows one to enhance significantly the hydrogen production in dedicated systems and the energy storage ability for the application in new generation batteries and other electrochemical technologies (i.e., thermoelectric pipes).

Acknowledgements

The publication was financed in the framework of the pro-quality program of the Lublin University of Technology: "Grants for financing the costs of interdisciplinary high-scoring publication" (Grant no. 5/GnG/2022). We want to express our gratitude to the researchers from the Department of Macromolecular Physics of Charles University in Prague, especially Prof. A. Choukurov, D. Nikitin, PhD, P. Pleskunov, PhD, K. Biliak, MSc, M. Protsak, MSc, and M. Tosca, MSc, for the fruitful technical support during the implementation of grant 5/GnG/2022, the introduction to the topic of electrical nanofluids, and numerous fruitful consultations and discussions on the topics undertaken.

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