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Multiple nanoparticles for improvement of thermal and dielectric properties of oil nanofluids

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Abstract: To improve the performance and increase the lifetime of oil-filled transformers, the thermal and dielectric properties of the transformer oil should be enhanced. Recently, nanotechnology was used as an effective science in the field of transformer oil development. In this study, barium titanate (BT) nanoparticles were inserted into the base transformer oil by a concentration of 0.005 g/L as an individual nanofluid sample (INFS). This insertion enhances the heat transfer coefficient by 33% but the breakdown voltage (BDV) was decreased by >10%. To overcome this problem of dielectric properties degradation, other three hybrid nanofluid samples (HNFS) were prepared using three different types of metal oxide (MO) nanoparticles; titania, alumina, and silica. These samples were prepared by adding a concentration 0.01 g/L of MO nanoparticles together with 0.005 g/L of BT nanoparticles into the oil. The thermal and dielectric properties of HNFS were measured to study the behaviour of nanoparticles hybridisation on transformer oil properties. HNFS using titania nanoparticles provided the best composition regarding either BDV or heat transfer coefficient. Dynamic light scattering (DLS) technique was used to evaluate the particle size distribution of hybrid nanoparticles and to clarify the corresponding physical mechanisms behind the obtained enhancement.

1 Introduction

In order to expand and upgrade the electrical power grid, the performance improvement of power transformers is indispensable. Oil-filled transformers are one of the popular types of power transformers. Transformer oil provides both thermal cooling and electrical insulation functions. So, improving its thermal and dielectric properties affects positively the whole performance of power transformer.

Recently, some researchers studied the effect of nanoparticles dispersion into the base insulating oil on its thermal and dielectric properties under the name of nanofluids. For the thermal properties of the insulating oil, Timofeeva *et al.* inserted a 5% volume fraction of SiO_2 nanoparticles into synthetic oil and could enhance the thermal conductivity by about 12% compared to the base insulating oil [1]. Mansour *et al.* concluded that the heat transfer coefficient of nanofluid was affected by the concentration of nanoparticles and the weight percentage of the surfactant [2]. In [3], it was concluded that thermal properties of the transformer oil based on cadmium sulphide quantum dots increased over that of pure transformer oil. Shukla *et al.* could improve the thermal conductivity of naphthenic transformer oil by 14.5% using the dispersion of 0.12% weight fraction of nanodiamond (ND) into the base oil [4]. The percentage enhancement continued to increase against temperature and attained about 17% at 120°C. In another research, ND of 0.1% weight fraction could enhance the thermal conductivity of mineral oil up to 10% at 30°C and 70% at 100°C [5].

Regarding dielectric properties of transformer oil nanofluids, Jin *et al.* presented that mixing of silica (SiO_2) nanoparticles with the base oil enhances the breakdown voltage (BDV) by about 27% compared to the base oil at humidity level of 20–30 ppm [6]. In [7], the dielectric properties of transformer oil were improved with the insertion of Fe_3O_4 nanoparticles. In [8], surface-modified alumina (Al_2O_3) and titania (TiO_2) nanoparticles were inserted to the base oil to increase the BDV against nanoparticles concentration until a certain level. The enhancement in dielectric properties was extended to aged transformer oil [9, 10], where adding TiO_2 nanoparticles could decrease effectively the dielectric dissipation factor and increase the breakdown strength of thermally aged oil.

Usually, metal oxide (MO) nanoparticles contribute effectively to the enhancement in dielectric properties, but have a limited impact on thermal properties. For example, SiO_2 nanoparticles did not enhance the thermal conductivity when added to mineral oil [11]. On the other hand, some nanoparticle types used for improving thermal properties have a limited impact on dielectric properties such as ND [4], and sometimes cause degradation in such properties such as carbon nanotube [12].

The aim of this study is using multiple nanoparticles for simultaneous enhancement of both thermal and dielectric properties of transformer oil. For thermal properties, the enhancement is usually based either on the thermal conductivity of nanoparticles themselves or on the random motion of nanoparticles and their collision with fluid molecules, which is denoted as Brownian motion [13, 14]. For oil-based nanofluids, usually the concentration of nanoparticles is small in order to keep stability. So, the contribution of nanoparticles themselves to the enhancement in thermal properties is limited. Also, small concentration cause large inter-filler distance making enhancement due to Brownian motion not effective. Accordingly, phonon transport can be considered the effective mechanism in enhancing thermal properties of oil-based nanofluids [15]. In this regard, barium titanate (BT) nanoparticles were selected as the first candidate in this study to enhance thermal properties of transformer oil, since they have sufficient number of phonons resulted from Ti-O-based lattice vibrations [16]. The second candidate in this study was selected among some of the effective nanoparticles that improved the dielectric properties of transformer oil. These nanoparticles are TiO_2, SiO_2, and Al_2O_3 nanoparticles.

First, BT nanoparticles were dispersed into the base transformer oil with a certain concentration to prepare an individual nanofluid sample (INFS). Heat transfer coefficient, relative permittivity, dissipation factor, electrical conductivity, and BDV were measured for this sample as a reference sample before hybridisation with another type of nanoparticles. Hybrid nanofluid sample (HNFS) was prepared using TiO_2, SiO_2, and Al_2O_3 nanoparticles. The same measurements were done for HNFS, and their properties were evaluated.

2 Experimental proceedings

2.1 BT nanoparticles preparation

In this study, BT nanoparticles were prepared by solid-state synthesis. The solid-state synthesis has multiple advantages. It is simple, environmental-friendly, and low-cost method. The solid-state method was based on solid-state reactions of mixed powders $Ba(NO_3)_2$, TiO_2, and tartaric acid at high temperatures. To follow this method in the current study, 0.1 mole of $Ba(NO_3)_2$, 0.1 mole TiO_2, and 0.3 mole of tartaric acid were grinded together for 2 h. The mixture was calcinated in an oven at 1000°C for 4 h, and then, was kept in a dry desiccator for storage.

The prepared BT sample has been characterised by several techniques to elucidate its structure and properties. Identification and characterisation of BT sample based on its diffraction pattern were obtained via XRD as shown in Fig. 1. The diffraction angle and intensity of the sample's peak are well consistent with that of the previous reported XRD of BT [17]. The morphology of sample structure and its average particle size was examined by high resolution transmission electron microscopy (TEM), as shown in Fig. 2. The TEM image confirmed that nanoparticles have nearly spherical shape with an average particle size of about 40 nm. The obtained BT nanoparticles were then used for the preparation of nanofluid samples as presented in the next section.

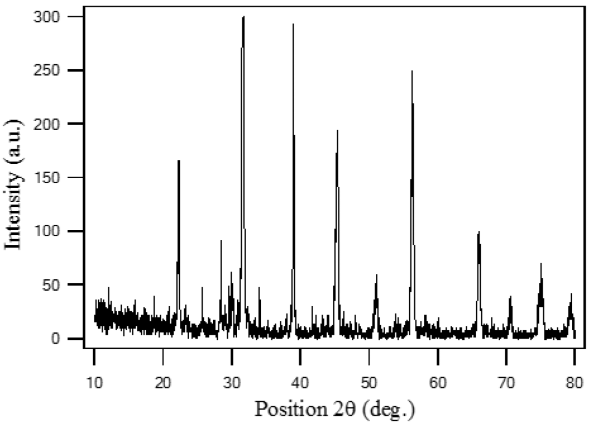

Fig. 1 *XRD of the prepared BT nanoparticles*

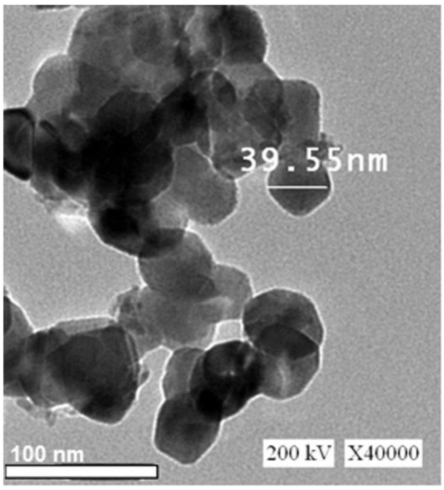

Fig. 2 *TEM image of the prepared BT nanoparticles*

2.2 Nanofluids preparation

The nanofluids were prepared by the dispersion of the nanoparticles into the pure insulating oil with a certain concentration. Tables 1 and 2 present the specifications of the transformer oil and the nanoparticles used in this study, respectively. As mentioned above, BT nanoparticles were prepared in the laboratory. MO nanoparticles were obtained commercially from Sigma Aldrich with the same average size for the sake of comparison. In this study, one INFS and three HNFS were prepared. In this study, no surfactant was used due to usage of different types of nanoparticles with different surface charging and, also, to avoid possibility of coiling up of surfactant chains covering BT nanoparticles and other types of nanoparticles. Instead, low concentrations of nanoparticles were used to avoid sedimentation of nanoparticles. For MO nanoparticles, 0.01 g/L or less without surfactant proved stable suspension [11, 18]. So, the concentration of 0.01 g/L was selected for TiO_2, SiO_2, and Al_2O_3 nanoparticles. For BT nanoparticles, its density is approximately two times of MO nanoparticles. So, their concentration was selected half of that used for MOs, i.e. 0.005 g/L.

The INFS consisted of a concentration of 0.005 g/L (BT/Transformer oil). To produce this sample, an amount of the base transformer oil was put in a calibrated beaker and the required weight of BT nanoparticles was evaluated using a sensitive electronic balance. Then, nanoparticles were inserted into the transformer oil and the beaker was placed on a magnetic stirrer with 1500 rpm for an interval of 15 min for the sake of mixing. For good dispersion of BT nanoparticles into the oil, ultrasonic irritation was used after the stirring process for 60 min by ultrasonic probe with an interval of 5 min per each 30 min. The produced sample was dried by a vacuum oven at 45°C for one day. This drying process acts towards removing gas bubbles and moisture content that produced during the ultrasonic irritation process. Fig. 3 summarises the preparation process of nanofluid samples. Before testing, each oil sample was cooled down in the surrounding air for an interval of 20 min.

Table 1 Insulating oil specifications used for nanofluids

Characteristic	Specification
manufacture	shell company
production name	Diala S2 ZU-I mineral oil
appearance	Clear and transparent
density at 20°C	0.875 g/cm^3
interfacial Tension at 25°C	0.045 N/m
flash point	140°C
pour point	−57°C
electric strength	> 60 kV
specific heat capacity	≈ 1.8 J/g.K
kinematic Viscosity at 40°C	9.4 mm^2/s
presence of oxidation inhibitor	Doesn't contain anti-oxidant additives

The HNFS is consisted of a concentration 0.005 g/L from BT nanoparticles and 0.01 g/L from other MO nanoparticles. To prepare these samples, an amount of the base transformer oil was calibrated by a graduated beaker. The required weight of BT nanoparticles was determined using a sensitive electronic balance. The BT nanoparticles were inserted into the base oil, and the beaker was put for 5 min on a magnetic stirring machine at 1500 rpm. Then, the sample was irritated by ultrasonic homogeniser for 5 min. During these 10 min, the required weight of MO nanoparticles was evaluated using the sensitive electronic balance. The MO nanoparticles were added to the mixture of BT nanoparticles and transformer oil. The sample was re-stirred for additional 5 min at the same rpm and irritated by ultrasonic probe for 60 min with an interval of 5 min after each 30 min. The subsequent drying process and testing are completely the same as carried out for INFS.

Table 2 Nanoparticles specifications used for nanofluids

Characteristic	Specification			
chemical compound	barium titanate	Titanium dioxide	silicon dioxide	Aluminum oxide
material form	powder	Powder	Powder	Powder
chemical formula	$BaTiO_3$	TiO_2	SiO_2	Al_2O_3
appearance	white crystal	white crystal	whitish yellow crystal	white crystal
odour	Odourless	Odourless	Odourless	Odourless
average grain diameter (nm)	< 50	20	20	20
density (g/cm^3)	6.02	4.23	2.65	3.99
specific heat capacity (J/gK)	≈ 0.5	≈ 0.7	≈ 0.7	≈ 0.9
thermal conductivity (W.m^{-1}.K^{-1})	6	4.8	1.3	12
electrical conductivity (S/m)	10^{-8}:10^{-10}	10^{-11}:10^{-16}	10^{-15}:10^{-19}	10^{-10}:10^{-16}
relative permittivity	1000:5000	10:85	3.6:4.2	7.8:11.1

2.3 Heat transfer coefficient of nanofluids

Transformer oil acts as not only an insulating medium but also a cooling medium for power transformers. So, good heat transferability is needed to get much cooling trace of the transformer oil. The calculation of the heat transfer coefficient for each prepared sample can be obtained by the following equation [2]:

$$H = \frac{Q}{T_i - T_o} \tag{1}$$

whereas H (W/m^2.K) is the heat transfer coefficient, Q (W/m^2) is the amount of heat transferred, T_i (K) is the temperature of the heater surface, and T_o (K) is the surrounding fluid temperature at the opposite side.

The experimental method that used for heat transferability test is presented in Fig. 4. The heater was connected with a controllable voltage source. Every 5 min, T_i and T_o were recorded until the occurrence of thermal steady state condition. After that, the heat transfer coefficient (H) could be evaluated as the average of the last six values.

2.4 Dielectric properties of nanofluids

The relative permittivity of any dielectric material relates directly to the dielectric polarisation intensity of this dielectric material. In oil/paper composite insulation system, the higher the relative permittivity of the oil the more uniformity the electric field at the oil/paper interface and the less the electrostatic charging. So, the increasing of the transformer oil relative permittivity is preferable. On the other hand, the dissipation factor is an important factor for the transformer oil, where it denotes to the dielectric losses inside the insulating oil, and thus, gives an indication to the total amount of contaminations. For each prepared sample, the relative permittivity (ε_r), the dissipation factor ($\tan\delta$) and the electrical conductivity (σ) were evaluated using Agilent E4980A Precision LCR meter. The measured circuit was adjusted in parallel mode of a capacitance (C_p) and resistance (R_p). Then, the above-mentioned parameters were obtained from the following equations:

$$\varepsilon_r = \varepsilon' - j\varepsilon'' \tag{2}$$

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'} \tag{3}$$

$$\varepsilon' = \frac{\mathrm{d}C_p}{\varepsilon_o A} \tag{4}$$

$$\varepsilon'' = \frac{d}{2\pi f \varepsilon_o \mathrm{A}R_p} \tag{5}$$

$$\sigma = 2\pi f \varepsilon_o \varepsilon'' \tag{6}$$

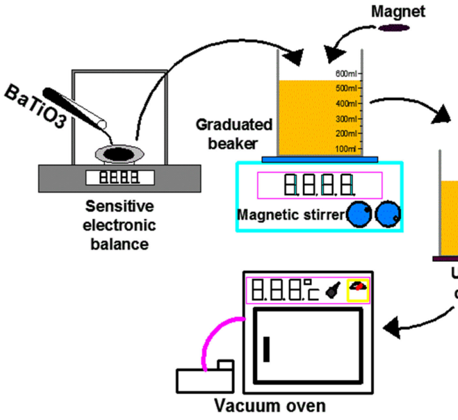

Fig. 3 *Nanofluid samples preparation process*

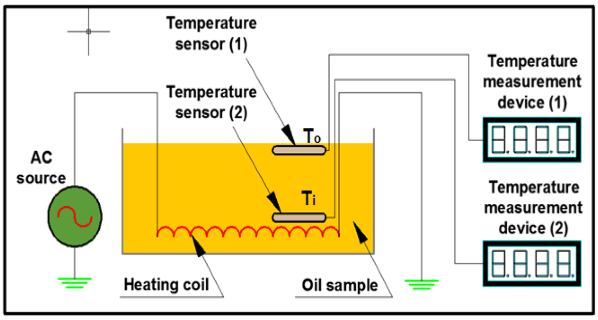

Fig. 4 *Experimental setup for heat transferability test*

whereas ε' is the dielectric coefficient, ε'' is the dielectric loss, d is the gap separation, A is the electrode cross sectional area, ε_o is the permittivity of free space and f is the frequency.

2.5 Dielectric strength of nanofluids

The BDV is the most essential feature of the insulating oil. Lower BDV causes adversely effects on the transformer performance. So, all prepared samples in this study were subjected to breakdown test using an oil tester. The test was performed according to IEC-60156 standard, in which the voltage ramp rate is adjusted at 2 kV/s, and the test electrodes were selected as a mushroom shape with gap spacing of 2.5 mm.

Ten measurements were recorded for each oil sample, and these values were analysed to construct the cumulative probability function of Weibull distribution. Weibull distribution analysis was provided to produce AC BDV at all probabilities with small number of tests [19]. If v is the BDV in kV, λ is the scale parameter in kV and ξ is the shape parameter, the cumulative probability function of Weibull distribution for neat insulating oil can be given by:

$$F(v) = 1 - \mathrm{e}^{-(v/\lambda)^{\xi}} \tag{7}$$

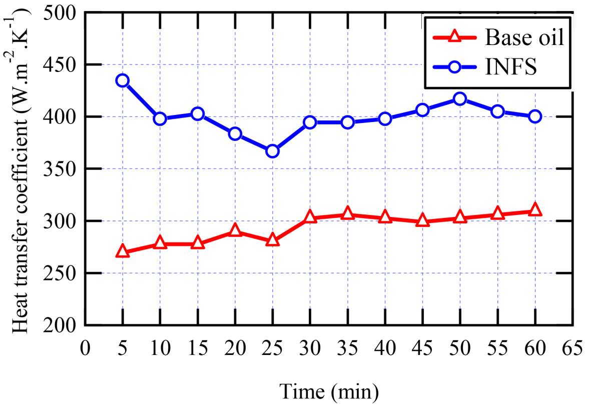

Fig. 5 *Heat transfer coefficient for INFS compared to base transformer oil*

Table 3 Heat transfer coefficient for INFS compared to base transformer oil

	Base oil	INFS
H (W.m^{-2}.K^{-1})	304	404
enhancing ratio (%)	—	33%

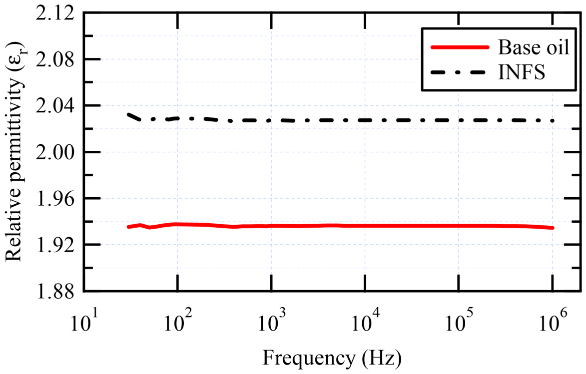

Fig. 6 *Relative permittivity for INFS compared to base transformer oil*

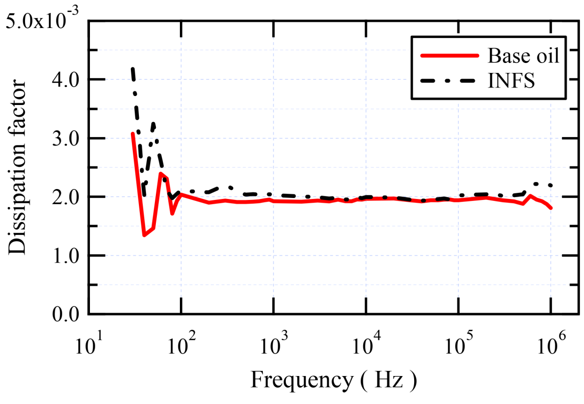

Fig. 7 *Dissipation factor for INFS compared to base transformer oil*

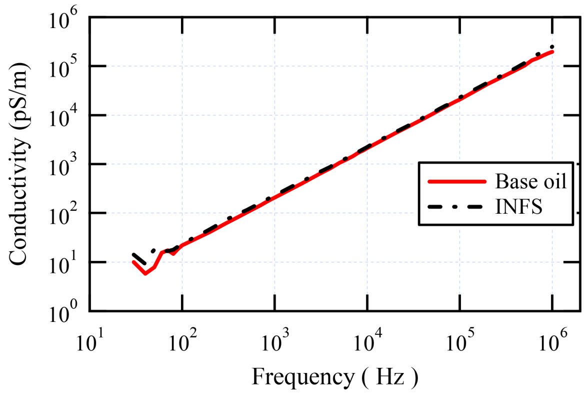

Fig. 8 *Electrical conductivity for INFS compared to base transformer oil*

Table 4 Obtained results of dielectric properties for INFS compared to base transformer oil at 50 Hz

Property	Base oil	INFS
relative permittivity	1.935	2.027
dissipation factor (× 10^{-2})	0.146	0.324
conductivity (pS/m)	7.84	18.3

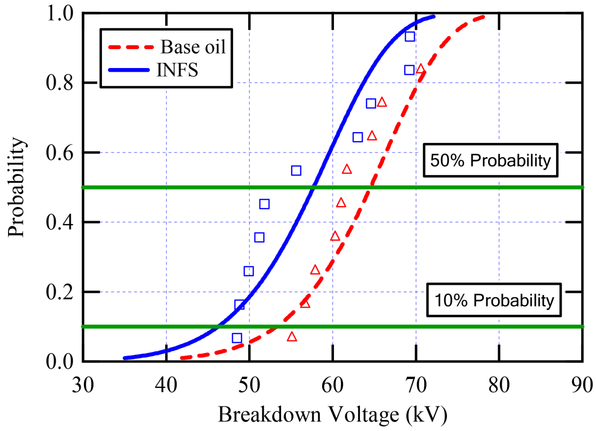

Fig. 9 *Cumulative probability versus BDV for INFS compared to base transformer oil*

Table 5 Weibull analysis for INFS compared to base transformer oil

Parameters	Base oil	INFS
scale parameter (kV)	67	60.24
shape parameter	9.8	8.5
v at 50% probability (kV)	64.5	57.7
v at 10% probability (kV)	53.3	46.3

3 Results and discussions

3.1 Individual nanofluids sample

3.1.1 Heat transferability: The heat transfer coefficient was evaluated for the base transformer oil sample and INFS as shown in Fig. 5. INFS provided a significant enhancement of the heat transferability by about 33% compared to as presented in Table 3.

3.1.2 Dielectric properties: The measured capacitance and resistance obtained from LCR meter in parallel mode were analysed to evaluate the relative permittivity, the dissipation factor and the electrical conductivity for each oil sample with frequency variation from 20 Hz to 1 MHz. Figs. 6–8 show the relative permittivity, the dissipation factor and the electrical conductivity for base transformer oil and INFS for the considered frequency range. For the power frequency of 50 Hz, the relative permittivity of the INFS was increased by about 5%, while the dissipation factor was doubled with a corresponding increase of electrical conductivity from 7.84 to 18.3 pS/m as summarised in Table 4. So, it is clear that using only BT nanoparticles has an adverse effect on dielectric losses in oil-based nanofluids.

3.1.3 Breakdown voltage: Fig. 9 shows the cumulative probability function for the INFS and that for the base transformer oil sample. Scale parameter, shape parameter, BDV at 50% probability and BDV at 10% probability are presented in Table 5. Due to the dispersion of 0.005 g/L BT nanoparticles into the base oil, the BDV at probabilities of 50 and 10% were decreased by about 10.5 and 13%, respectively, compared to that of the base transformer oil.

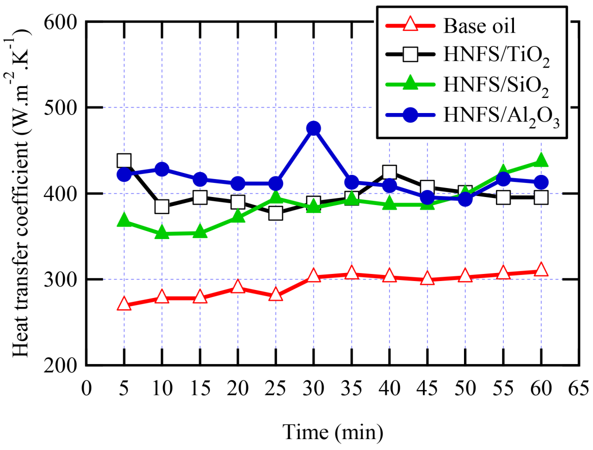

Fig. 10 *Heat transfer coefficient for all HNFS compared to base transformer oil*

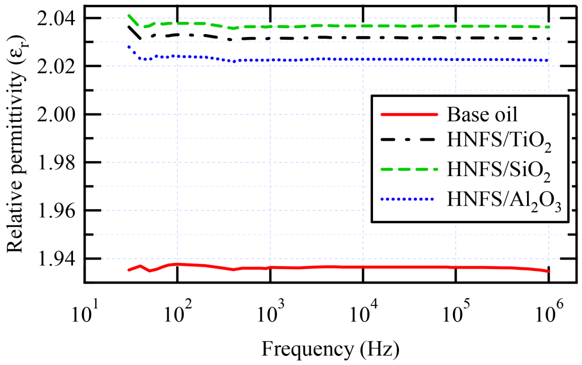

Fig. 11 *Relative permittivity for all HNFS compared to base transformer oil*

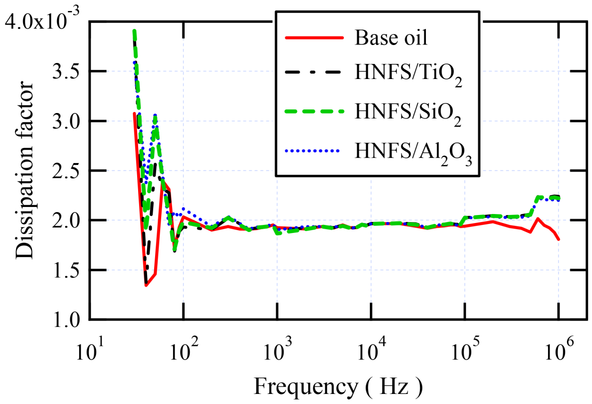

Fig. 12 *Dissipation factor for all HNFS compared to base transformer oil*

3.2 Hybrid nanofluids samples

3.2.1 Heat transferability: There are three prepared HNFS. These samples were prepared as mentioned above with TiO_2 nanoparticles, SiO_2 nanoparticles and Al_2O_3 nanoparticles. For each HNFS, the heat transfer coefficient was evaluated as shown in Fig. 10 and summarised in Table 6. From the obtained results, it is clear that the enhancement of the heat transferability due to the HNFS is very close to that obtained with INFS.

3.2.2 Dielectric properties: LCR meter in parallel mode provided the measurement of each capacitance and resistance for each HNFS. These measurements were analysed to evaluate the relative permittivity, the dissipation factor and the electrical conductivity for each HNFS with the same range of frequency as used for INFS. Figs. 11–13 show the relative permittivity, the dissipation factor and the electrical conductivity, respectively, for all HNFS with the mentioned frequency range. At frequency of 50 Hz, these results are summarised in Table 7. The relative permittivities of all HNFS were in the same range of INFS. The dissipation factor and the electrical conductivity decreased compared to INFS. The lowest dissipation factor and lowest conductivity were obtained for HNFS with TiO_2 nanoparticles.

Table 6 Heat transfer coefficient for all HNFS compared to base transformer oil

Sample type	H (W.m^{-2}.K^{-1})
base transformer oil	304
HNFS/TiO_2	403
HNFS/SiO_2	404
HNFS/Al_2O_3	406

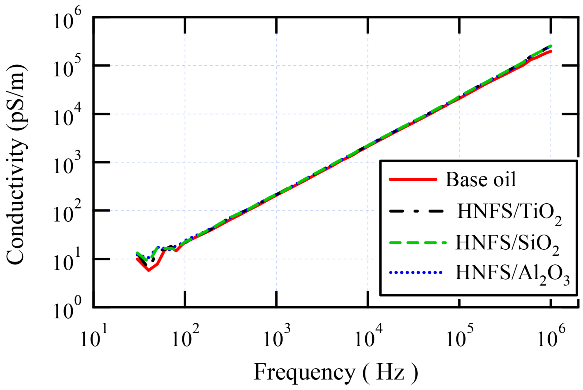

Fig. 13 *Electrical conductivity for all HNFS compared to base transformer oil*

Table 7 Obtained results of dielectric properties for all HNFS compared to base transformer oil at 50 Hz

Property	Base oil	HNFS/TiO_2	HNFS/ SiO_2	HNFS/ Al_2O_3
relative permittivity	1.935	2.03	2.03	2.02
dissipation factor ($\times 10^{-2}$)	0.146	0.256	0.303	0.308
conductivity (pS/m)	7.84	14.5	17.2	17.3

3.2.3 Breakdown voltage: Fig. 14 shows the cumulative probability function for all HNFS. Scale parameter, shape parameter, BDV at 50% probability, and the BDV at 10% probability are presented in Table 8. It is noted that, HNFS with TiO_2 nanoparticles has the highest BDV followed by that with SiO_2 nanoparticles. While, HNFS with Al_2O_3 nanoparticles exhibited BDV lower than that of the base oil. The percentage enhancements in BDV at 50% when using TiO_2 and SiO_2 nanoparticles were 42.9% and 18.6%, respectively, while the percentage decrement when using Al_2O_3 nanoparticles was 4.5%. So, HNFS with TiO_2 nanoparticles is considered the best composition either regarding dielectric losses or dielectric strength.

4 Physical mechanisms

Regarding the thermal properties, adding BT nanoparticles into the insulating oil has the main positive impact on thermal properties. This is attributed to transport of phonons resulted from Ti-O-based lattice vibrations [16]. Addition of any other types of MO nanoparticles did not exhibit a considerable improvement over the BT nanoparticles as evident from the results presented in Tables 3 and 6.

Regarding dielectric properties, INFS achieved an increase in the relative permittivity of the oil. This is attributed to the high dielectric constant of BT nanoparticles, which contributes positively to the increase in the dielectric coefficient of the

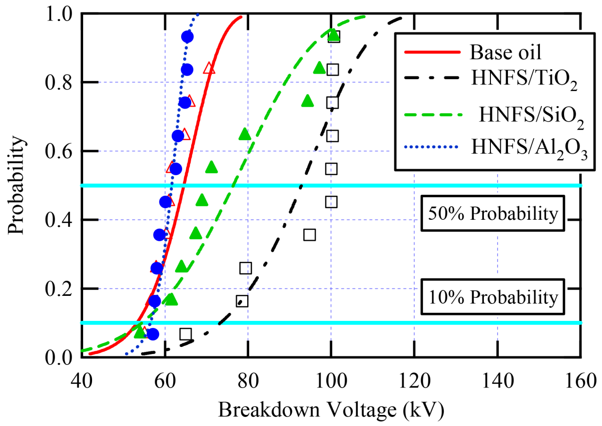

Fig. 14 *Cumulative probability versus breakdown voltage for all HNFS compared to base oil*

Table 8 Weibull analysis for all HNFS compared to base oil

Parameters	Base oil	HNFS/TiO$_2$	HNFS/SiO$_2$	HNFS/Al$_2$O$_3$
scale parameter (kV)	67	97.2	81.7	62.7
shape parameter	9.8	7.98	5.55	21.7
v at 50% probability (kV)	64.5	92.2	76.5	61.6
v at 10% probability (kV)	53.3	73.3	54.5	56.5

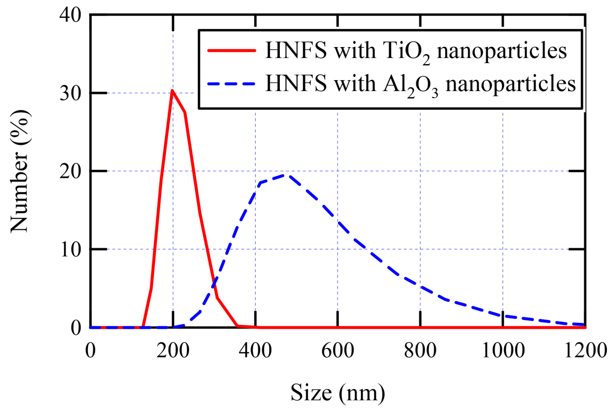

Fig. 15 *Particle size distribution of HNFS with different types of MO nanoparticles*

mixture. For dissipation factor and conductivity, they increased with adding BT nanoparticles due to two main effects. The first effect is the conductivity of BT nanoparticles themselves, which is high. The second effect is the number of charge carriers in the fluid, which increases due to the large difference in permittivity between BT nanoparticles and oil.

For HNFS, there is a difference in obtained results among different types of MO nanoparticles considered in this study. The HNFS prepared with TiO_2 nanoparticles exhibited the highest BDV and the lowest dielectric losses and conductivity compared to that prepared with SiO_2 or Al_2O_3 nanoparticles. The second ranking of enhancement was obtained with SiO_2 nanoparticles, while, there is no significant enhancement with Al_2O_3 nanoparticles. To clarify this, the role of nanoparticles surface charging has to be considered. MO nanoparticles have different surface charging tendency based on their isoelectric points in relative to pH value of the oil that is measured around 7. SiO_2 and TiO_2 nanoparticles have negative surface charging tendency, while, Al_2O_3 nanoparticles have positive one. Regarding BT nanoparticles, their isoelectric point is 3 [20]. Thus, their surface charging is negative similar to that of SiO_2 and TiO_2 nanoparticles. So, when adding hybrid nanofillers of BT nanoparticles and such types they repulse against each other and keep dispersed. While, when adding BT nanoparticles with Al_2O_3 nanoparticles, they attract to each other and become agglomerated. To confirm this, dynamic light scattering (DLS) analysis was done for HNFS prepared with TiO_2 nanoparticles and Al_2O_3 nanoparticles. The results are depicted in Fig. 15. It is clear that HNFS prepared with TiO_2 nanoparticles has smaller particle size than that prepared with Al_2O_3 nanoparticles, in spite of the initial size of all MO nanoparticles is the same. So, as a general rule for nanoparticles hybridisation in nanofluids, they should have similar surface charging polarity. The further enhancement of TiO_2 nanoparticles over SiO_2 nanoparticles attributed their ability to attract electrons [21] and trap them into shallow traps [22].

5 Conclusions

This paper studied the effect of using hybrid nanoparticles of BT and MO for simultaneous enhancement of thermal and dielectric properties. Heat transfer coefficient, relative permittivity, dissipation factor, and BDV were measured for all samples. From the obtained results, the following comments can be considered:

i. The dispersion of 0.005 g/L BT nanoparticles into transformer oil improved the heat transferability by about 33%, although, the dissipation factor and BDV were degraded.
ii. The dispersion of 0.005 g/L BT nanoparticles with 0.01 g/L TiO_2 nanoparticles improved the heat transferability by about 33% and increased the BDV by about 43%. Also, the degradation in dissipation factor was significantly alleviated.
iii. The enhancement was obtained only when using BT nanoparticles with TiO_2 and SiO_2 nanoparticles. This was explained in terms of isoelectric point and surface charging of nanoparticles.
iv. Based on DLS analysis, it is confirmed that hybrid nanoparticles should have the same surface charging polarity for effective dispersion and favour impact on thermal and dielectric properties.

6 Acknowledgment

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