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Performance and limitation of mineral oil-based carbon nanotubes nanofluid in transformer application

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Abstract Transformer oil-based carbon nanotube (CNT) nanofluids which have unique dielectric behaviour, is effective as the posterity insulation fluids that can boost the performance of the transformer as they proposed inspiring, distinctive behaviour compared to existing transformer oil which is widely used in practice namely mineral oils. With this motivation, the effect of AC breakdown voltages for two sonication duration (30 min and 120 min) techniques were applied in producing nanofluids, two different diameter sizes of CNTs (< 8 nm and > 20 nm) and five different weight concentrations (0.01 g/L to 0.2 g/L) are investigated. The results indicate CNT with a longer sonication process, a smaller diameter and low concentrations of CNT provides the highest breakdown values that gave a huge potential impact on the conventional transformer oil. The Weibull and Normal distributions functions are used in this paper to obtain a successful forecast of the lowest, average, and highest possibility of breakdown rates (1%, 50% and 90%). It figures out that, CNT nanofluid can reach the greatest breakdown efficiency as good insulating oil at 0.01 g/L concentration. To understand the characterization of CNT nanofluids samples in detail, Raman spectroscopy analysis, storage modulus, viscosity and heat flow of mineral oil have been evaluated accordingly as a function of increasing temperature.

1. Introduction

The transformer is considered as one of the most significant parts in the electricity network serving the transmission and distribution of electricity in the high voltage three-phase electrical power grid to the low voltage grids. Fundamentally,

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the transformer is an apparatus that converts electrical energy from one electrical circuit to another through mutual (electromagnetic induction) without alteration in frequency. While in operation, the transformer generates a lot of heat from resistive losses in the electrical and magnetic components that must be dissipated to keep them running safely. Once transformer heat increases critically, it will cause huge failure to the transformer and cost millions of dollars to replace and months to repair. Bartley has collected thousands of transformer's failures information to identify the causes of transformer failures and figure out that insulation is highly the main cause of deterioration in transformers [1]. Insulation is responsible for providing better quality and performance of transformer during its operation. Proper utilization of the insulation materials can determine the life cycle and capability of the transformer. Insulation system in transformer consists of either liquid or gas along with solid insulator materials depending on the type of transformers used in the industry. The dry-type transformer uses air as a cooling medium, while the liquid-type uses oil. Most transformers all over the world install liquid-filled type transformers due to the biggest advantage of the units; handle higher ratings, are cost-effective and are highly suitable for outdoor locations.

The liquid contains in the transformer, which acts as an insulator known as transformer oil. The main function of transformer oil is to prevent electric current between conducting elements and transfer the heat generated by active parts (magnetic core & winding) to the tank wall of transformers. Apart from that, transformer oil also suppresses corona and arcing and serves as a coolant in the meantime protecting the transformer. One of the required qualities in transformer oil is to maintain its dielectric stability at various temperatures for an extended period with excellent electrical insulating properties and suitable cooling ability. For decades, petroleum-based mineral oil has been used as a cooling medium or known as transformer oil in these types of transformers because of its chemical stability, low viscosity, higher pour point, high electric field strength, low dielectric losses and good long-term performances [2]. Mineral oil contains different types of hydrocarbon molecules such as aromatics C_nH_n (benzene C_6H_6), paraffin C_nH_{2n+2} (hexane C_6H_{14}), naphthenic C_nH_{2n} (cyclohexane C_6H_{12}) and others [3]. Although mineral oil has been used for a century, it is known to have low fire point temperature (low thermal characteristics) which can cause a major risk of fire during operation at very high temperatures.

Concerning fire safety and environmental issues, the development of high thermal conductivity of transformer oil for critical application is indispensable. One of the developments is by dispersion of nanomaterials in conventional transformer oil or known as nanofluid with aim of improving thermal characteristics of the insulating oil as well as enhancing the electrical properties of insulating oil. Fig. 1 represents the number of publications that studied nanofluid as alternative transformer oil from the year 2010 until January 2022 based on IEEE Xplore Digital Library. It seems that the awareness of nanofluid research as alternative transformer oil has risen as the year rises until 2019. However, the amount of literature on nanofluid studies has dramatically decreased from 35 to 15 publications (8 journals and 7 conferences). This could be because researchers are beginning to overcome the challenges of working with nanofluids, particularly in the study of stability and thermophysical behaviour.

In 2018, Abd-Elhady et al. [4] reviewed the electrical breakdown voltages performances of nanofluids based ZrO_2 nanoparticles. According to these researchers, it has been proven that this mixture has great potential in a practical application through experimental measurements. Compared to conventional transformer oil value, the nanofluids produced a 202.60% increment in breakdown voltage performance, which means that ZrO_2 has a great contribution toward transformer oil in future. Furthermore, Radja et al. [5] also claimed that SiO_2 also can enhance the breakdown voltage and possible help reclamation of aged transformer oil. Before proceeding to fill the fresh transformer oil in a transformer or routine maintenance, the breakdown voltage test or known as dielectric strength test is usually included among the quality controls and acceptance tests for transformer oil. In general, the term breakdown voltage is the maximum electric field that transformer oil can withstand under ideal conditions without failure. The higher breakdown produces, the better an electrical insulator it makes.

Other than ZrO_2 and SiO_2, there were also studies on nanofluid based CCTO [6], Fe_3O_4 and Al_2O_3 [7]. However, very restricted information was available concerning electrical breakdown performances of carbon nanotubes (CNT), which has a distinctive structure and remarkable mechanical, thermal, and electrical characteristics. The CNT has a nano-sized particle, which is 10,000 times smaller than human hair. These attributes enable CNT to be readily dispersed or absorbed into the component of insulation [8]. This paper is therefore encouraged to explore the impact of CNT in conventional transformer oil (petroleum-based mineral oil) by referring to the IEC standard processes. Hence, the primary goal of this job is to perform an experimental investigation on the efficiency of AC breakdown performance or dielectric strength of mineral oil-based CNT nanomaterial at distinct sonication duration, different diameter sizes of CNT (<8 nm and >20 nm) and various concentrations of nanofluids, respectively. To observe the modification caused by CNT in oil, Raman spectroscopy analysis, dynamic mechanical analysis and simultaneous thermal analysis are also studied in this paper.

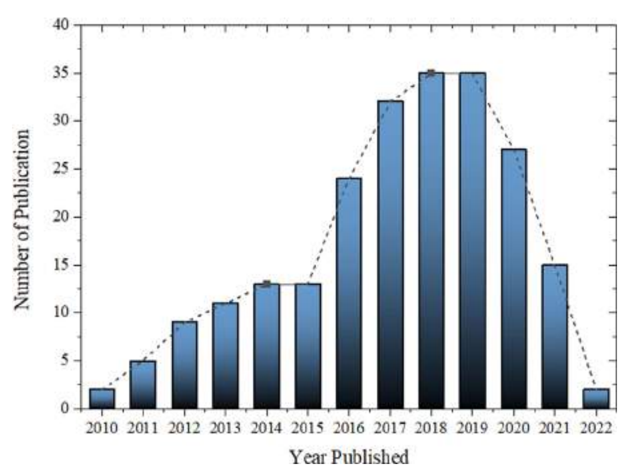

Fig. 1 Number of publications regarding nanofluid as alternative transformer oil from the year 2010–2019.

2. Preparation of CNT nanofluids

Generally, there are two primary techniques to prepare nanofluids, which are the one-step and two-step process. In the one-step process, nanoparticles are synthesized in the base fluid by chemical method, while in two-step process firstly prepares nanoparticles in a form of powders and then dispersed in base fluid [9]. However, the two-step method is extensively used by most researchers due to the production method being found straightforward and much cheaper. Hence, a two-step preparation method is carried out in preparing CNT nanofluids in this research. Fig. 2 shows the block diagram of the two-step preparation process of nanofluids. Firstly, CNTs were mixed and stirred with mineral oil by using a magnetic stirrer at 520 rpm for 30 min to speed the reaction and improve the composition of samples. Then, the most significant process is carried out by dispersing CNTs in fluid through sonication process using sonicator with 700 W of power rating, 20 kHz capability, 10% amplitude set up, 5 s timing cycle on and 3 s off interval for two duration (30 min & 120 min). To reduce the humidness contained and unwanted gas in CNT nanofluids, which may lead to the degradation of oil quality, the samples were dried and degassed at the temperature of 85 °C in a vacuum oven with pressure less than 5 Mbar for 48 h direct. The moisture contained in the fluids can release charge carriers and therefore decreases the dielectric strength. After the nanofluids samples have been dried and degassed, the samples were cooled at ambient temperature (20 to 25 °C) over 24 h under vacuum conditions [10]. Before any testing, the moisture content of each prepared sample was measured in the range of ~25 ppm using a moisture metre. Nanofluids are not types of mixtures ready to use, which can lead to variability in thermal conductivity and heat flow characteristics depending on the preparation process. Different types of nanomaterial and fluids possibly have a different approach on sonication parameters. The detail of commercial mineral oil as based transformer oil is shown in Table 1, while Fig. 3 and Table 2 depicts the SEM images of CNTs used in this study, respectively.

Table 1 Detail of petroleum-based mineral oil.

Parameter	Specification
Appearance	Clear & Bright
Kinematic Viscosity at 40 °C	9.8 cSt
Kinematic Viscosity at −30 °C	1120 cSt
Pour point	−55 °C
Water content	20 ppm
Density at 20 °C	0.8830 g/ml
Dielectric Dissipation Factor at 90 °C	0.0002
Acidity	0.0005 mgKOH/g
Flashpoint	145 °C

3. Experimental setup

The AC breakdown voltage tests measure the electrical stress, which insulating oil can withstand by referring to IEC 60156 standard procedures. This test is conducted using a DTA 100C BAUR oil breakdown voltage test equipment along with a 400 ml glass test vessel as shown in Fig. 4, whereas Table 3 shows the equipment's technical data. The AC voltage applied to the electrodes and voltage power frequency will gradually enhance until the oil breaks down and there will be a spark between the electrodes. The standard specifies the use of mushroom/spherical shaped electrodes and the optional use of stirring impeller/magnetic bead stirrer/ no stirring in the testing procedures.

The IEC 60156 [11] recommends that a separate glass test vessel be used for each different type of oil sample and the vessel should be rinsed at least three times to ensure that there is no contamination exists during the test. The oil samples also need to pour swiftly into the test vessel with minimum turbulence to avoid air trapped in the test vessel. The voltage is continuously applied at the electrodes with a 2 kV/s rising rate with 2 min initial standing time and another 2 min interval with stirring action after each breakdown occurs. A total of 50 readings of AC breakdown results was taken for each experiment in this study.

In this work, a Raman spectrometer (Renishaw InVia Raman Microscope) is used to analyse the chemical composition and energy band gap of the oil compound, as detailed in section 5.1. It is a highly successful approach for determining the precise bonding structure status of oil samples under various situations. For experiment setup, 785 nm laser excitation sources with 1200 lines/mm grating, 10 s exposure period, and 1% laser power are employed in this investigation. Section 5.2 focuses on measuring viscosity and storage modulus using a Perkin Elmer DMA 8000 to measure the viscoelastic characteristics of CNT nanofluids from 30 °C to 90 °C. Throughout the experiment, the DMA 8000 is configured with a clamping system without interrupting the temperature profile. A few drops of oil samples are placed on the material pocket tool (52.5 mm × 12.8 mm × 8.0 mm) that is specifically designed for DMA 8000. The material pocket is a stainless stell envelope that holds the sample so it can be mounted in a DMA equipment. Section 5.3 employs the PerkinElmer Simultaneous Thermal Analysis (STA 6000) and differential thermal analysis to determine the sample weight change and heat flow (DTA). This equipment offers temperature control ranging from 15 °C to 1000 °C, a quick cool-down period, and exact monitoring, making it ideal for research applications.

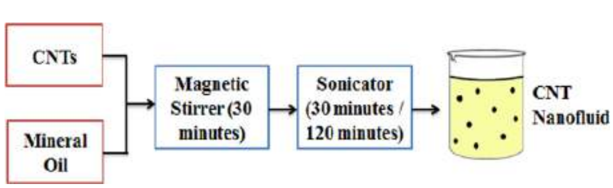

Fig. 2 Two-step preparation process of nanofluid.

4. AC breakdown voltage measurements

4.1. Sonication duration

Sonication is a de-agglomeration process to break up CNTs and promote dispersion into base oil to gain better stability of nanofluids. This process is usually employed for preparing the nanofluids mixture samples and is highly dependent on

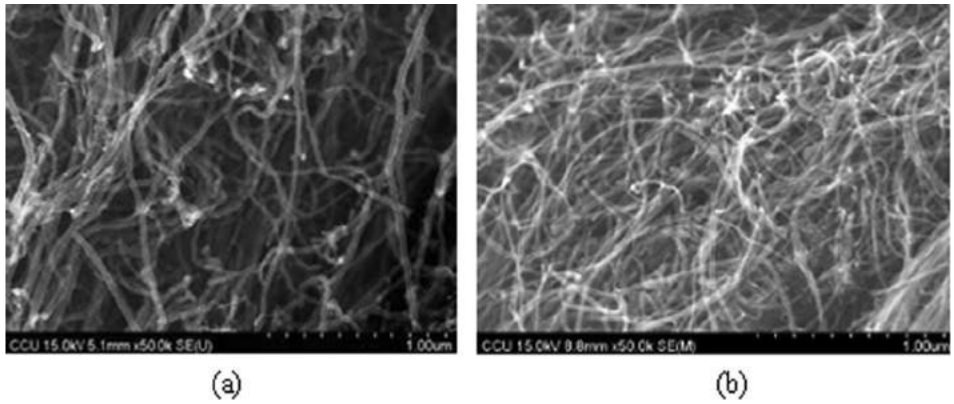

Fig. 3 Scanning Electron Microscopy (SEM) of (a) CNT with < 8 nm and (b) CNT with > 20 nm.

Table 2 Detail of Carbon Nanotube (CNT).

Parameter	Sample 1	Sample 2
Outer Diameter	< 8 nm	20–30 nm
Inner Diameter	2–5 nm	5–10 nm
Length	10–30 µm	10–30 µm
Purity	95%	95%
Supplier	Chengdu Organic Chemical Co. Ltd., Chinese Academy of Science	Chengdu Organic Chemical Co. Ltd., Chinese Academy of Science
True density	~2.1 g/cm^3	~2.1 g/cm^3
Making method	Chemical Vapor Deposition	Chemical Vapor Deposition

Fig. 4 DTA 100C BAUR oil breakdown voltage tester with 400 ml test vessel in accordance with IEC 60156 standard.

sonication duration. Two durations of sonication, (30 min and 120 min) are selected in this study to determine the most stable nanofluids generated. Colloidal suspensions were formulated by CNTs with average particle diameter size < 8 nm at a range of 0.01 g/L to 0.05 g/L weight concentrations. Fig. 5 reported the comparison of AC breakdown voltages between three ranges (0.01, 0.03 and 0.05 g/L) of CNT nanofluids.

Table 3 Technical Data of BAUR Breakdown Voltage Tester.

Parameter	Value
Input voltage	90–264 V (50/ 60 Hz)
Output voltage	0–100 kV_{rms} symmetrical
Degree of protection	IP 32
Voltage slew rate	0.5–10 kV/s
Switch-off time	< 10 µs
Accuracy	0–100 kV ± 1 kV

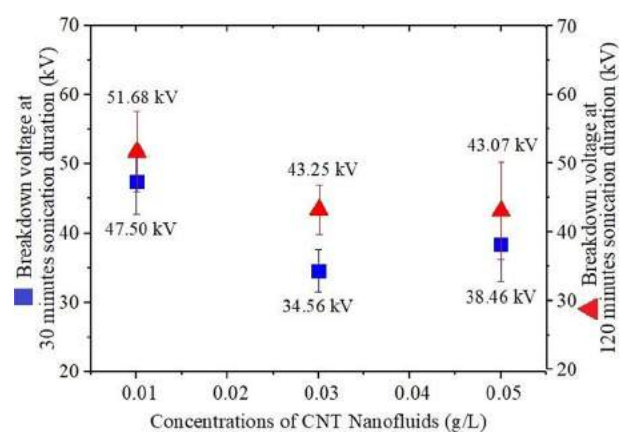

Fig. 5 Average of AC breakdown voltages of CNT nanofluids for two duration of sonication (left: at 30 min; right: at 120 min).

Based on Fig. 5, after 120 min of the sonication process, all three levels of concentrations of CNT nanofluids achieve greater breakdown voltage than 30 min of sonication. There are improvements of 8.80% (0.01 g/L), 25.14% (0.03 g/L) and 11.80% (0.05 g/L) at a longer period of sonication. The 30 min of sonication duration did not provide sufficient dispersing energy to overcome the agglomeration of CNTs as the CNTs condition remains clustered. CNT nanofluids with 120 min of sonication duration received better energy to disperse the CNT uniformly, lose the agglomeration which resulted in stable colloid suspension, and produce higher AC breakdown voltages value. This result is also consistent with the findings of Farade et al. [12] who studied the effect of sonication for 10, 20, 30 and 60 min on graphene-based nanofluids. The researchers reported that sonication for 60 min resulted in higher absorbance, more homogeneous dispersion and stable suspension, which all contributed to the enhancement of AC breakdown voltages. According to research, nano-sized particles are significantly more homogeneous than micro-sized particles due to the vigorous Brownian motion [13–15].

However, at 120 min of sonication, there are slightly decrement trend is observed as concentrations increase. When the duration of the process is longer and the input energy intensity is higher, it is possible to achieve better dispersion quality. Such aggressive treatment might also adversely affect the nanotube's structure. Consequently, deteriorate the electrical breakdown voltage properties of nanofluid. Kole et al [16] investigated the measurement size of ZnO nanofluid and discovered that the time of sonication could contribute to the rise in particle size. This is due to the presence of cavitation caused by the sonicator, which is attributed to the substantial shear force resulting in nanoparticle aggregation [14]. However, different types of materials have diverse chemical bonding structures and stability mechanisms. There are still no specific standards on the proper procedure to prepare nanofluids using the sonication method, especially on the duration of the sonication process. Yu Zhen et al. [17] have a strong opinion that a suitable procedure for producing nanofluids will result in more stable and better performance nano-transformer oil.

4.2. Diameter sizes of carbon nanotube

CNT is a long and thin cylindrical shape that is unique in terms of its shape and size, but they also have remarkable physical properties. Pokryvailo et al. [18] studied that the performance of CNT is directly related to the diameter sizes of the CNTs nanomaterial. Conductivity and non-conductivity of CNT are usually based on the CNT's degree to twist and diameter dimensions. In this study, the performance of AC breakdown voltages at two CNT diameter sizes (<8 nm and >20 nm) are evaluated after selecting the length of the sonication duration for 120 min. Fig. 6 demonstrates a comparison of AC breakdown voltage performances at two distinct CNT diameter sizes for three concentrations of CNT nanofluids (0.01 g/L, 0.03 g/L and 0.05 g/L).

Fig. 6 shows that CNT's with diameter sizes of $<$ 8 nm obtain greater breakdown voltage values compared to larger diameter sizes. Seems that at 0.01 g/L CNT nanofluids (<8 nm) achieved the highest breakdown voltage as much as 51.68 kV among all samples where there is 70.50% enhancement observed compared to CNT nanofluids with >20 nm diameter size at a similar concentration. However, these results are inconsistent with a report [19], where the author figure out that increasing the size of Fe_3O_4 nanoparticles will enhance the capability of breakdown performance of nanofluids and significantly influence the increment of electrical potential well depth. The depth of the electrical potential well of nanofluids clarify the influence of nanoparticles on electron trapping and is related to the improvement of breakdown performance. These differences may be due to the distinct structure between

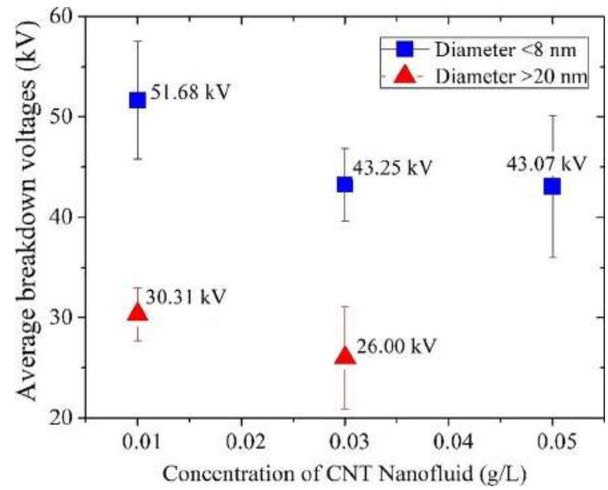

Fig. 6 Average of AC breakdown voltages of CNT nanofluids at two diameter sizes.

Fe_3O_4 and CNT, which consist of a two-dimensional hexagonal lattice of carbon atoms as a form of a hollow cylinder.

It is noted that the breakdown tester was unable to measure the value of AC breakdown voltage for >20 nm diameter size at 0.05 g/L concentrations. This is due to the range of output voltage generated outside the tolerance range. This occurrence might be due to the instability of 0.05 g/L CNT nanofluid that causes the AC breakdown tester to disable to detect the measurements of the oil sample. This also could be due to CNT's characteristic that might have converted from non-conductive materials to conductive properties at 0.05 g/L concentrations.

4.3. Various concentrations

The AC breakdown voltage measurements have been recorded and compared at five different concentrations to analyse the quality of oil samples before and after adding some CNTs. Fig. 7 illustrates the obtained results and the corresponding

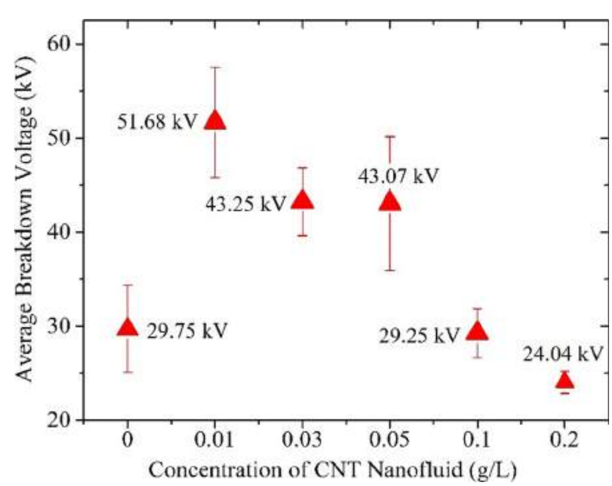

Fig. 7 Average of AC breakdown voltages of CNT nanofluids at various concentrations ranging from 0 to 0.2 g/L.

Table 4 Parameter of CNT nanofluids at various concentrations.

Oil Samples	Concentration (g/L)	Average (kV)	Standard Deviation (kV)	Standard Error	Minimum (kV)	Maximum (kV)
Mineral Oil	0.00	29.75	1.1873	0.6567	21.2	40.1
CNT Nanofluids	0.01	51.68	5.8473	0.8269	36.5	61.9
	0.03	43.25	3.6032	0.5096	35.4	49.9
	0.05	43.07	7.0953	1.0034	28.4	54.3
	0.1	29.25	2.5988	0.3675	22.3	33.5
	0.2	24.04	1.1873	0.1679	21.6	26.9

parameter are provided in Table 4. The standard deviations measured in all types of oil samples were within the range below 7.00 kV. Hence, the data is considered valid since the readings are concentrated around the mean value. While the standard error is a way to know how close the average of a reading of breakdown voltage is to the average of the whole measurements. The standard error values for nearly all samples are less than one based on Table 4, which implies that the 50 measurements of breakdown voltages for each sample are consistent.

Noted in Fig. 6, the breakdown voltage reaches the maximum value at 0.01 g/L concentration followed by 0.03 g/L and 0.05 g/L CNT nanofluid. It can also be observed that the breakdown voltage tends to decrease as concentrations level rise. However, compared to the reference voltage value of mineral oil, 0.01 g/L, 0.03 g/L and 0.05 g/L CNT nanofluids produced higher breakdown voltage, as much as 73.71%, 45.38% and 44.77% while for 0.1 g/L and 0.2 g/L CNT nanofluids, the breakdown voltage is lower compared to the mineral oil. The differences between 0.2 g/L CNT nanofluid and mineral oil were slightly low (−19.19%). The slight decrement may be due to the instability of CNT nanofluids, which tends to form aggregations and clusters. These results were found similar to Purbarun et al., study where the authors observed that the breakdown voltages tend to decrease after 0.025 wt% of CNT [20]. On the basis of electron trap theory and the experimental results produced, it can be concluded that fast-moving free charges can be slowed down by the adsorption of free charge by CNT as shown in Fig. 8(a), which is in line with theory proposed by Zahn et al. [21].

Therefore, streamer propagation is obstructed and cause the breakdown voltage performance of the proposed nanofluid to improve. A nanotube can be good insulating oil if there is a large bandgap between the conduction band and valence band [22–26]. However, CNT is also possible to transport electrons and act as a conduction bridge which leads to an abrupt decrease in the breakdown performance of nanofluid as shown in Fig. 8(b). This viewpoint is supported by Sami et al. [27], who discovered that the distortion of the electric field caused by accumulated charges is responsible for the decrement performance of electrical breakdown. However, the diameter, concentration, amount of twist in CNT's lattice determines whether it is obstructing or assisting electrons. At lower concentrations, the distortion in the electric field can be ignored, allowing charge trapping to take precedence and an increase in breakdown voltage to occur. However, at higher concentration levels, the higher distortion in the electric field enhances the breakdown propagation of the streamer that leads to earlier breakdown.

4.4. Statistical analysis of experimental data

Reliability of the insulating material under certain electrical stress is necessary for industrial design purposes and this parameter is calculated by a few distributions that suit the data best. To estimate the probability of breakdown failure, a total of 50 breakdown voltage measurements were obtained and analysed using appropriate statistical techniques; as it presents the condition of failure, maintenance decisions, considering diagnostic measurement and life data. To confirm the results of breakdown voltages, two types of distribution have been used in this study; a distribution that fit the data most based on Anderson-Darling goodness-of-fit and Normal distribution, which is the most common distribution used in statistical

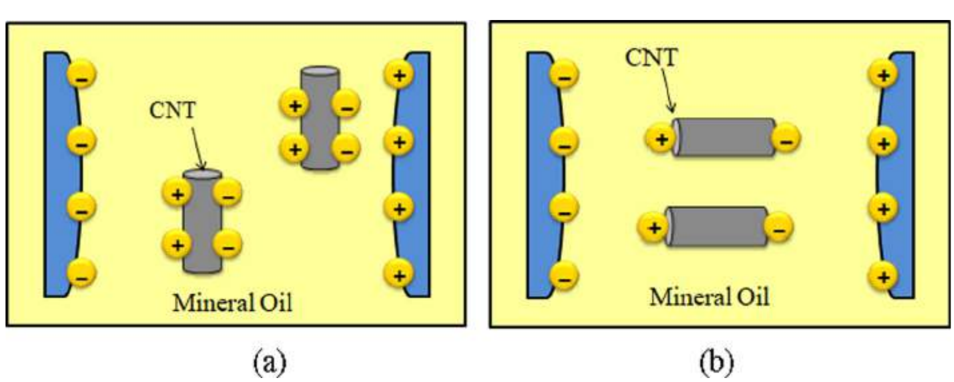

Fig. 8 Charge transport model for CNT nanofluids where: (a) CNT as a charge barrier. (b) CNT as a conduction bridge.

Table 5 Goodness-of-fit ranking based on Anderson-Darling.

Ranking	Mineral Oil	CNT Nanofluids				
		0.01 g/L	0.03 g/L	0.05 g/L	0.1 g/L	0.2 g/L
1	Lognormal 0.27954	Weibull 0.27369	Weibull 0.25879	Lognormal 0.67711	Weibull 0.45654	Gamma 0.17861
2	Gamma 0.29133	Gamma 0.80479	Gamma 0.71863	Gamma 0.69511	Gamma 0.77933	Lognormal 0.19143
3	Weibull 0.9459	Lognormal 0.9338	Lognormal 0.77874	Weibull 0.94502	Lognormal 0.83277	Weibull 0.67906
4	Exponential 4.8066	Exponential 9.1217	Exponential 6.676	Exponential 6.0787	Exponential 9.4065	Exponential 6.0727

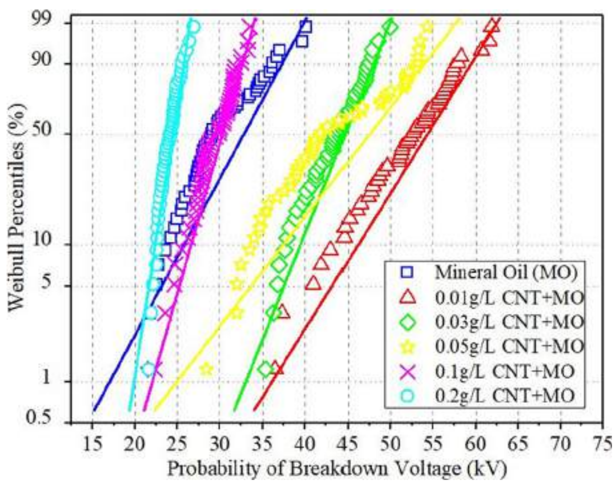

Fig. 9 Probability breakdown voltages of samples using Weibull Distribution.

analysis based on histogram pattern. Generally, many types of distribution can be used to calculate the probability of breakdown failure. In this study, four different types of distribution are selected which are Weibull distribution, log-normal distribution, exponential distribution and gamma distribution [28]. Table 5 shows the goodness-of-fit test distribution ranking based on the Anderson-Darling method.

Referring to the data in Table 5, the analysis results show a better fit in three types of distributions, which are Weibull, gamma, and lognormal distributions while exponential distribution returns the lowest ranking in the Anderson-Darling goodness-of-fit test. However, since most of the oil samples are well suited with the Weibull distribution, hence, the Weibull distribution used in this study to estimate the expected breakdown voltage value for mineral oil and five concentrations of CNT nanofluids. Furthermore, the differences between Weibull and other ranking were slightly small.

The solid lines in Fig. 9 represent the Weibull fitting of 50 breakdown voltage results and the graph depicts that 0.01 g/L CNT nanofluid rank the highest probability of breakdown voltage as its line shifted to the rightest side while the highest concentration produces least favourable. The parameters and correlation coefficient of the Weibull distribution plot is shown in Table 6. The parameter α is the shape parameter, also known as the Weibull slope, which gives the Weibull distribution its flexibility while β is the scale parameter, which determines the range of distribution.

In this paper, generally, there were three significant probability distribution percentiles were identified based on Weibull fitting in Fig. 9, which are 1%, 50% and 90% as shown in Table 7 [29]. From the information, it is noted that 0.01 g/L CNT nanofluid has achieved the highest rank of breakdown voltage among the samples. Furthermore, the failure rate for all concentrations of CNT nanofluids at 1% probability of breakdown voltage achieved better breakdown voltage compared to mineral oil. At the standard failure rate used by industry, which is 50%, it is observed that only 0.1 g/L and 0.2 g/L CNT nanofluids show lower breakdown voltage compared to mineral oil. For 0.2 g/L CNT nanofluid, the probability of breakdown voltage of 0.2 g/L CNT nanofluid is decreased around 19%, while 0.1 g/L CNT nanofluid is only

Table 6 Parameter and Correlation of Weibull Distribution Plot based on Fig. 9.

Parameters	Mineral Oil	0.01 g/L	0.03 g/L	0.05 g/L	0.1 g/L	0.2 g/L
α	6.89	10.94	14.61	7.00	13.86	21.06
β	31.76	54.16	44.84	46.07	30.39	24.60

Table 7 Breakdown probability at 1%. 50% and 90% percentiles.

Breakdown Probabilities (%)	AC voltage Breakdown (kV)					
	Mineral Oil	CNT Nanofluids				
		0.01 g/L	0.03 g/L	0.05 g/L	0.1 g/L	0.2 g/L
1	16.29	35.57	32.75	23.87	21.80	19.77
50	30.12	52.36	43.74	43.72	29.59	24.18
90	35.84	58.41	47.47	51.92	32.25	25.50

1.76% compared to mineral oil. Based on the overall outcomes, CNT nanofluid can regard as a potential candidate to replace conventional transformer oil because it exhibits improvement in electrical properties although at the lowest possible breakdown failure rate.

The normal distribution is one of the most common distributions used in statistics and calculating the probabilities of breakdown voltages of dielectrics. It also describes how the values of a variable are distributed and for this paper, histogram and normal distribution is applied to detect eventual anomalies in the distribution of breakdown voltage data in different ranges of voltages. Fig. 10(a) until Fig. 10(f) give the histograms of the distribution of breakdown voltages for mineral oil and CNT nanofluids from 0.01 g/L to 0.02 g/L

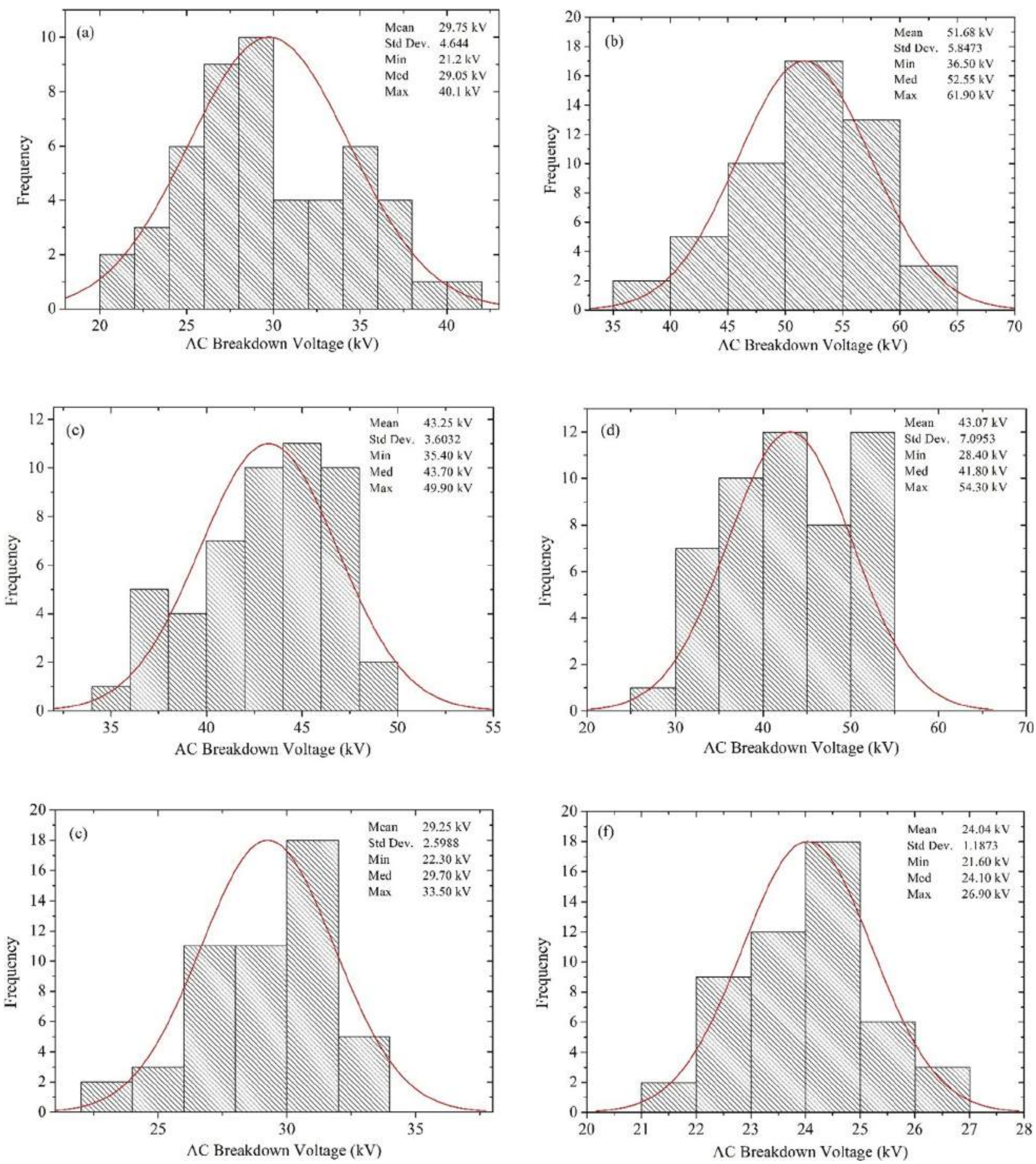

Fig. 10 Histogram of AC breakdown voltages measurements for (a) mineral oil, (b) 0.01 g/L CNT nanofluid, (c) 0.03 g/L CNT nanofluid, (d) 0.05 g/L CNT nanofluid, (e) 0.10 g/L CNT nanofluid and (f) 0.20 g/L CNT nanofluid.

concentrations. These representations graphically summarize and visualize the distribution of the breakdown voltage data set. To evaluate the problem with the non-normality of distribution, one can refer to skewness and kurtosis. Skewness is the measurement lack of symmetry, while kurtosis represents the measurement shape of the distribution, where the ideal symmetrical data should be zero for perfect distribution. Table 8 gives the value of skewness and kurtosis for the investigated samples.

Table 8 Skewness and Kurtosis of breakdown voltages of mineral oil and CNT nanofluids.

Oil Samples	Skewness	Kurtosis
Mineral Oil	0.29472	−0.52807
0.01 g/L CNT Nanofluid	−0.65103	0.17516
0.03 g/L CNT Nanofluid	−0.42564	−0.6717
0.05 g/L CNT Nanofluid	−0.01337	−1.08681
0.1 g/L CNT Nanofluid	−0.56302	−0.06148
0.2 g/L CNT Nanofluid	0.14047	−0.32787

Observed from the skewness value, all the patterns of counts or known as frequency distributions of breakdown voltage data is symmetric to the left and right of the centre point. As for kurtosis, which describes the degree of peakedness of distribution, it appears that most of the CNT nanofluids based on Table 8 are getting results with less than 1 and greater than −1 except for 0.05 g/L CNT nanofluid. However, kurtosis higher than 3 does not exclude the data of normality. Hence, according to the value, consider that all of the investigated samples obey the normal distribution.

5. Characterization of samples

Raman spectroscopy, Dynamic Mechanical Analysis (DMA), and Simultaneous Thermal Analysis (STA) are used to characterize prepared CNT nanofluids. Fig. 11 shows the optical microscope image captured by Raman for mineral oil,

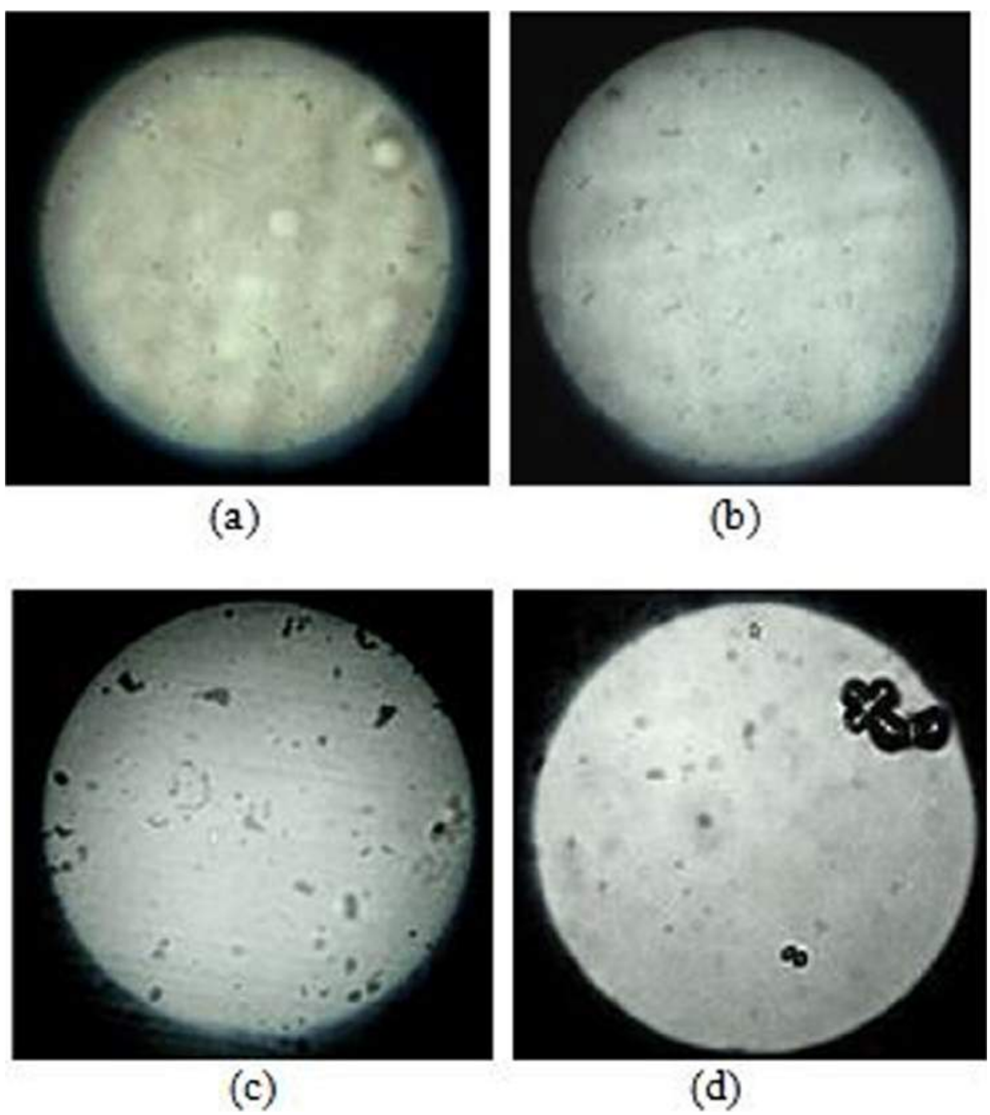

Fig. 11 Optical microscope images of (a) mineral oil; (b) 0.01 g/L CNT nanofluid; (c) 0.03 g/L CNT nanofluid; (d) 0.05 g/L CNT nanofluid.

0.01 g/L, 0.03 g/L and 0.05 g/L CNT nanofluids with 5 μm focus.

5.1. Raman spectroscopy analysis

As shown on the microscope image of CNT nanofluid, concentrations of 0.01 g/L show a uniform dispersion of CNT in mineral oil, while at 0.03 g/L and 0.05 g/L, the existence of CNT's aggregates in the mineral oil specimens can be seen. Mineral oil containing CNT have obvious round and black shape particles indicating the undistributed of CNTs. It also can be observed that at 0.05 g/L CNT concentration, the sample contained large size black tubes attached. This might be due to the duration of sonication processes or sound energy applied to agitate particles were not enough to disperse the CNTs in the mineral oil sample. Besides, as the concentration of CNT rises, the agglomerations also increased. The agglomeration of CNTs in mineral oil may be one of the mechanisms that induced CNT nanofluids to break down, which explains the results of this study. The agglomerations not only can cause settlement and clogging of micro-channels but also reduce the overall effective thermal conductivity of nanofluids. Therefore, researchers have looked for many other options to avoid aggregation problems such as adding some surfactants or dispersants to improve the immersion of nanomaterials in the based oil. Some of them are using surface modification techniques to modify the chemical, physical characteristics and surface structure of nanomaterials [30]. Furthermore, the appropriate selection of nanoparticles, amount of weight or volume concentrations and compatibility of nanoparticles with based oil are also attributed to the stability of nanofluid [31].

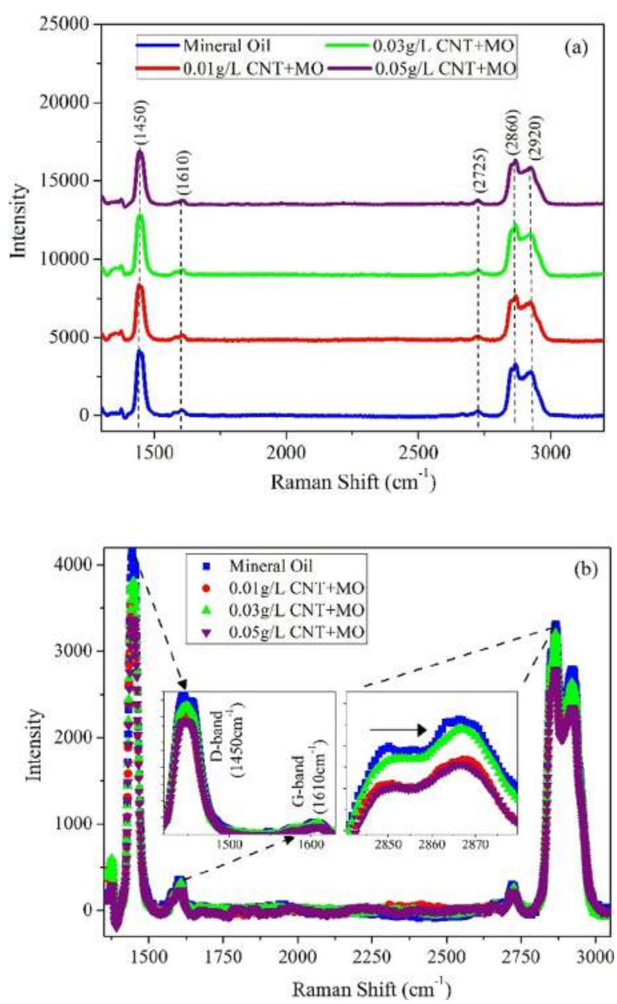

Fig. 12 Raman spectra of mineral oil and CNT nanofluids between (a) 1150 cm-1 to 3100 cm-1 and (b) first and second-order region.

Raman spectroscopy equipment is a standard non-destructive tool to confirm the amorphous behaviour of the CNT nanofluids and mineral oil. It is a very effective method to characterize the detailed bonding structure of samples. Fig. 12 graphically depicts the Raman spectra measurements of CNT nanofluids from 1350 cm^{-1} to 3100 cm^{-1} Raman shifts. No additional peaks associated with impurity were found as all the samples showed numerous features, which related to complex oil structures and agreed well with Somekawa [32]. Based on Fig. 11(a), the large peak centred at 1450 cm^{-1} and 1610 cm^{-1} corresponds to C=O stretching and C=C stretching vibration modes. Noted that, there is the presence of kerogen structure in samples, which is characterized by first-order region (1100–1800 cm^{-1}) and the second-order region (2200–3400 cm^{-1}) as shown in Fig. 11(b) vibrational bands [33]. The most prominent features in Raman spectra are D-band and G-band, where D-band is caused by the disordered structure of graphene existing in the sample, while G-band is referred to as graphitic structure. Observed that, 0.05 g/L CNT nanofluid has the lowest D-band compared to mineral oil. Peak reduction might indicate less crystallinity or improper arrangement of atoms in the lattice. In the higher wavenumber side, the large peak at between 2700 and 3100 cm^{-1} region shows C-H stretching mode which represents the diversity of CH, CH_2 and CH_3 group [34,35]. Referring to Raman spectra of the second-order region, it is analyzed that the Raman peak is shifting towards a higher wavenumber as concentrations increase, which is related to the shorter bond length of C-H in oil molecules due to the doping effect. Based on the overall observation of Raman shift, there was no additional peak existed in disposed transformer oil chemical structure after adding MWCNT which means that MWCNT

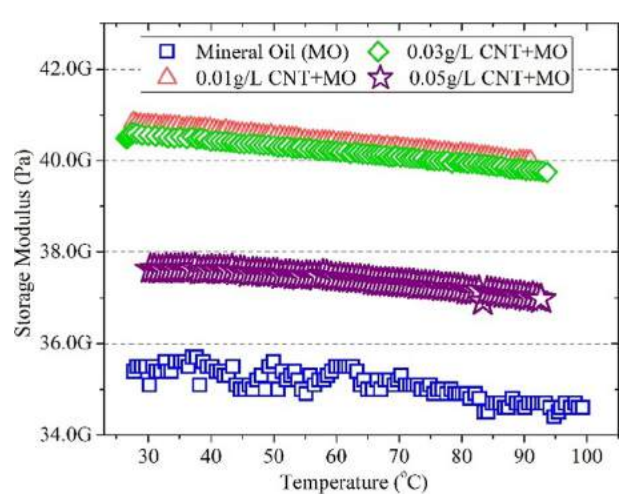

Fig. 13 The storage modulus of oil samples at various temperature.

molecules did not disrupt the structure behaviour of transformer oil.

5.2. Dynamic mechanical analysis

Dynamic mechanical analysis or known as DMA is a thermal analysis method that measures the viscoelasticity and other significant properties of materials. The DMA equipment can mimic the operating conditions of the material at the desired temperature. In this study, DMA measured and compared the storage modulus, which represents the energy dissipated, and dynamic viscosity of mineral oil, 0.01 g/L–0.05 g/L CNT nanofluids as shown in Fig. 13 and Fig. 14.

Observed that, mineral oil sample has the lowest storage modulus compared to other samples. It was also seen that when the temperature rose, the storage modulus of CNT nanofluids dropped gradually while the mineral oil pattern reacts differently. However, the storage modulus of mineral oil was constantly stable at 30–40 °C and began to fluctuate until the temperature of mineral oil reach 70 °C. There are possibilities that some voids and pores appeared during sample processing which caused slight changes in the mineral oil. At normal ambience temperature, 0.01 g/L and 0.03 g/L CNT nanofluids were able to store higher electrical energy charges compared to other oil samples. The greater stored charge indicates the high permittivity of dielectric materials. In particular, increase amount of CNT up to 0.03 g/L indicate higher particle-particle interaction. Furthermore, CNT nanofluids also have a more consistent storage modulus and are correlated with breakdown voltage results. In this study, the correlation of dynamic viscosity and temperature of mineral oil and CNT nanofluids with various concentrations was experimentally measured and the result is shown in Fig. 14.

Dynamic viscosity is another important parameter to be considered for good alternative transformer oil. The force required by a fluid to overcome its internal molecular friction for the fluid to flow is defined as dynamic viscosity. In other words, dynamic viscosity is defined as the tangential force per unit area required to move a fluid in one horizontal plane with respect to another with a unit velocity while the fluid's molecules remain a unit distance apart [36]. The natural convection flow is very dependent on the viscosity of insulating oil [2]. The dependence of viscosity of mineral oil and CNT nanofluids on temperature was a manifestation of its cohesive and thermal energies behaviour. Based on Figure 14, 0.01 g/L concentration has the highest viscosity, followed by 0.03 g/L, 0.05 g/L and mineral oil. These results explained the behaviour of breakdown voltage in this study. Furthermore, looking at the results, mineral oil showed an inconsistent pattern of viscosity while CNT nanofluids consistently show a decrease in viscosity along with the temperature. It is normal for every liquid sample to become less viscous at a higher temperature. With the rise of temperature, the attraction between the polar molecules dropped while their thermal energies increased. Thereby, the viscosity of oil samples also decreased. As the viscosity of oil samples increases, the dielectric properties of insulating oil samples decreases [37] thus affecting the reliability of the transformer. This is due to the impact of thermal expansion, where an increase in temperature implies an increase in kinetic energy (fast motion). This occurrence leads to weaker storage modulus along with viscosity at a higher temperature.

5.3. Simultaneous thermal analysis

To examine the effect of the heat flow of the samples, the Simultaneous Thermal Analysis (STA) 8000 is used to test the 20 to 40 mg of mineral oil and CNT nanofluids samples. The STA sensor is a temperature sensor, which is calibrated from 30 °C to 100 °C to generate data for heat flow with an accuracy of ±5% or better. Fig. 15 shows the comparison of heat flow between mineral oil and three varied concentrations of CNT nanofluids.

Heat flow represents the amount of heat that samples can transfer from a hotter system to a cooler system in a particular area. Based on Figure 15, 0.01 g/L achieves the highest amount of heat flow followed by 0.03 g/L, 0.05 g/L and mineral oil with increasing temperature. Seems that mineral oil has the lowest amount of heat transfer, which means mineral oil could increase its temperature rapidly and then lower the breakdown performance. Besides, lower heat flow can cause degradation

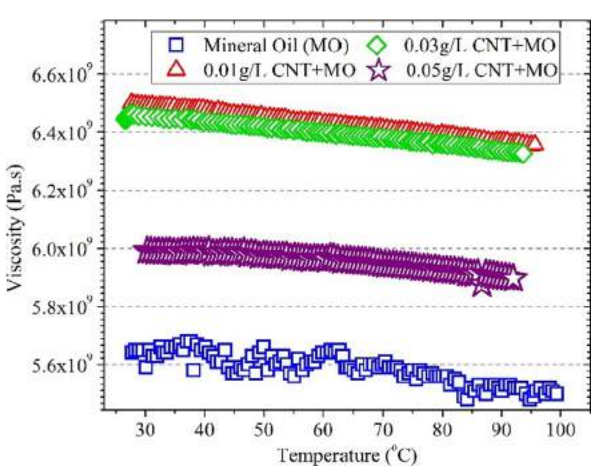

Fig. 14 The dynamic viscosity of oil samples at various temperature.

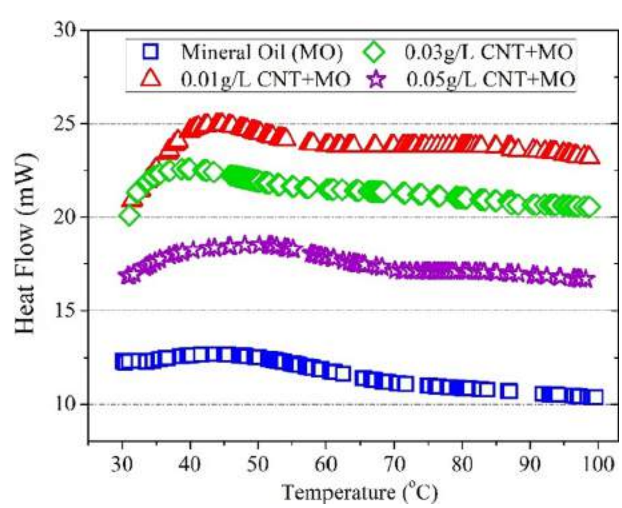

Fig. 15 Heat flow of oil samples at various temperature.

over time and equipment malfunctions while higher heat flow provided high performance and stability in the system. Therefore, this study, proves that at 0.01 g/L concentration nanofluid is regarded as the most appropriate convective heat transfer medium relative to mineral oil. Good convective heat transfer medium help minimize the losses produced, known as copper or I^2R losses and core or iron losses that affect the reliability of the transformer.

6. Conclusion

This paper explores the idea of adding trace amounts of CNTs into typical mineral oil with the purpose of breakdown voltages enhancement for a longer lifetime of the transformer. In this study, the patterns of AC breakdown voltages based on various factors have been taken into consideration. These involve different sonication duration processes, sizes of nanomaterial, and various concentrations of CNTs ranging from 0. 01 g/L–0.2 g/L. Looking at the results, it was observed that the sonication process of nanofluids within 120 min by dispersing < 8 nm CNT diameter sizes at 0.01 g/L weight concentration gave the highest breakdown voltage value. It produced the most stable suspension of nanofluids, which inhibit the movement of electrons in the oil samples.

This occurrence might be due to the behaviour of CNT that could trap and avoid electrons from being transported from one electrode to another and causing breakdown performance to improve significantly. However, for more than 0.1 g/L weight concentration the behaviour of CNT begins to alter as a conduction bridge and assist electrons to travel fast between electrodes, which causes breakdown voltage to decrease. The reliability of insulation under certain electrical stress is calculated in this paper using Weibull distribution, which was selected through the Anderson-Darling goodness-of-fit analysis.

Based on the distribution, the concentrations up to 0.05 g/L CNT nanofluid, breakdown voltages performances unexpectedly improved compared to the reference mineral oil. Furthermore, CNT nanofluid showed up to 118.35% enhancement of the breakdown voltage at 1% probability (the lowest possibility of breakdown voltage) for 0.01 g/L CNT nanofluid. Moreover, at 1% breakdown probability, all amounts of CNT's concentration achieved better breakdown voltage compared to reference mineral oil used in this study. It can be concluded that adequate preparation of nanofluids, diameter sizes of CNTs and number of concentrations could have such an enormous impact on the efficiency of electrical breakdown performance in the implementation of transformer oil. Furthermore, based on a few testings, shown that the 0.01 g/L CNT nanofluid has a remarkable impact on the storage modulus, viscosity and thermal behaviour that contributes to the breakdown performance. Higher storage modulus indicates higher energy storage capability of the material. Nevertheless, 0.01 g/L CNT nanofluid is characterized by decreasing pattern of viscosity with increasing temperatures. Because of decreasing viscosity values, the thermal performance (by convection) of oil samples will be efficient for transformer application. However, extensive research needs to be performed to analyze the charge accumulation and determine its suitability for long term operation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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