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The Effect of Interfacial Zone Due to Nanoparticle–Surfactant Interaction on Dielectric Properties of Vegetable Oil Based Nanofluids

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ABSTRACT Nanoparticles are generally anti-lipophilic and have a tendency to aggregate when they are embedded in nanofluids. Thus, surfactants have a major role in achieving long term dispersion stability of nanofluids through surface modification of nanoparticles. However, the surfactant changes the structure of the interfacial zone around nanoparticles and can have a crucial impact on the dielectric properties of dielectric nanofluids. Accordingly, this paper aims to clarify the role of interfacial zone on dielectric properties of vegetable oil based Al_2O_3 nanofluids including relative permittivity, dissipation factor, and AC breakdown voltage. Moreover, a polarization model of nanofluids was proposed to calculate their relative permittivity considering surfactant effect. Different filler levels ranging from 0.01 wt% to 0.05 wt% were used and various temperature ranges were considered to validate the proposed model. Relative permittivity calculated from the proposed model was almost in line with experimental results. Proposed model depicted that surfactant contributes for orientational polarization of nanoparticles in the relative permittivity calculation. So, improvement in the relative permittivity of dielectric nanofluids has been attained from 0.01 wt% to 0.05 wt% of Al_2O_3 nanoparticles due to both internal and orientational polarization of nanoparticles. Regarding dissipation factor of prepared nanofluids, it decreased against the weight percentage of nanoparticles, while the mean AC breakdown voltage increased against the weight percentage. All these effects were discussed considering the structure of interfacial zone and its impact on the alignment of oil chains and the energy loss of electrons.

INDEX TERMS Al_2O_3 nanoparticles, surfactant, relative permittivity model, nanofluids, dielectric properties.

NOMENCLATURE

2a Diameter of spherical nanoparticle
CSO Cottonseed oil

d Surfactant thickness
DBF Dielectric base fluid
DNF Dielectric nanofluid
dQ Charge on nanoparticle's small volume
 E_0 External electric field applied in the direction of x-axis

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k	Boltzmann constant
N_1	Number of base oil molecules/unit volume
N_2	Number of nanoparticle molecules/unit volume
NP	Nanoparticle
p	Electric dipole moment of nanoparticles
SDBS	Sodium Dodecylbenzene Sulfonate
T	Temperature
TBHQ	Tertiary Butylhydroquinone
V	Volume of each nanoparticle
α_1	Polarizability of base oil molecules
α_2	Inner polarizability of NPs
α_3	Orientalional polarizability of charged NPs
ϵ_0	Permittivity of vacuum
ϵ_{NF}	Relative permittivity of DNF
ϵ_1	Relative permittivity of base fluid
ϵ_2	Relative permittivity of NPs
φ_{NP}	Volume fraction of NPs in DNF
σ_1	Charge density of NPs without surfactant polarization
σ_2	Charge density of NPs with surfactant polarization

I. INTRODUCTION

Power transformers can be classified as small, medium and large power transformers based on their ratings. Power transformers are generally liquid cooled and use billions of litres of transformer oil. Distribution transformer units are more numerous and therefore consume more transformer oil than other power transformers. The demand for transformer oil is increasing every day. The increase in energy consumption is a key factor in the increasing demand for transformer oil. Mineral oils are the conventional transformer oil those are currently used in the majority of transformer applications. Environmental concerns and the non-renewable nature of mineral oils shifted paradigm to vegetable oil dielectrics [1], [2].

With the advancement in nanotechnology, the concept of nanofluids has been widely developed to improve various properties of the base fluids. For dielectric fluids, the constituted nanofluids can be denoted as DNFs. Recent studies showed that stably dispersed nanoparticles into DBFs improves dielectric properties of DNFs to a considerable extent [3]–[9]. These improvements are attributed to repeated trapping and de-trapping of charge carriers into the fluid causing a limit in the transportation of these charge carriers under the influence of electric field. In the continuous media these traps are generally made by chemical defects [10], in case of natural esters these traps are due to carbonyl groups. In DNFs, additional trapping sites are formed by NPs due to the difference in the relative permittivity of NPs and that of base oil [11].

Several models have been proposed to calculate the relative permittivity of these composite fluids. In this regard, the Maxwell-Garnett model was widely used in the description and measurement of the relative permittivity of DNFs [12], but this model is not considered being sufficient

since it is just a function of the concentration and relative permittivities of the constituents. As a result, a model was constructed in [13] based on S-parameter retrieval and considering the scattering of nanoparticles. The S-parameter retrieval model could estimate more accurate relative permittivity than that obtained from the Maxwell-Garnett model. Moreover, a proposed model based on NP polarization was proposed to investigate the relative permittivity of composite fluids [14], and could exhibit more closer results to the experimental ones. However, all these models [12]–[14] did not consider the surfactant polarization.

NPs are generally anti-lipophilic and have a tendency to aggregate when they are embedded in nanofluids. Thus, using of surfactants became a major process in achieving long term dispersion stability of nanofluids through surface modification of nanoparticles [15], [16]. In [17], the effect of surfactant amount on stabilization of nanoparticles were clarified and it was concluded that excess amount of surfactant can hinder stability due to the formation of double surfactant chains around the surface of nanoparticles. This concept was effectively utilized in [18], where the usage of suitable surfactant amount could enhance the dispersion behavior of TiO₂ nanoparticles into transformer oil with a positive impact on the breakdown strength. The usage of surfactant with various ultrasonication periods were investigated in [19] and it was found that the longer the ultrasonication period up to optimized period, the higher the UV-Vis absorbance and the more homogenous the dispersion of nanoparticles.

Regarding the effect of surfactant on long term stability, it was discussed in [20] and [21]. In [20], aluminium nitride nanoparticles were modified with two layers of surfactants before dispersion into the transformer oil. This modification enabled to keep the stability of the prepared nanofluids more than 6 months with an enhancement up to 7% in thermal conductivity and up to 50% in positive lightning impulse breakdown. In [21], the oleic acid was used as a surfactant with iron oxide nanoparticles to fill the transformer oil and prepare what is called magnetic nanofluids. The stability of these magnetic nanofluids was measured over 7 years.

In addition to the role of surfactant in keeping long term stability, it was found that it plays an important role in strengthening the interfacial zone around nanoparticles. This interfacial zone is considered to have a major impact on the dielectric properties of nanofluids [22], like its significance in solid nanodielectrics as reported by many researchers [23]–[25]. Since surfactants are frequently used to keep long term stability and to strengthen the interfacial zone, it is crucial to consider the surfactant polarization in the relative permittivity calculation of nanofluids.

This research focuses on to clarifying the role of interfacial zone on dielectric properties of CSO based Al₂O₃ nanofluids. The effect of surfactant polarization on relative permittivity of CSO based Al₂O₃ nanofluids is also clarified. Cottonseed oil is a non-edible vegetable oil that represents a promising candidate to replace conventional mineral oil. Detailed characterization of Al₂O₃ nanoparticles and methodology

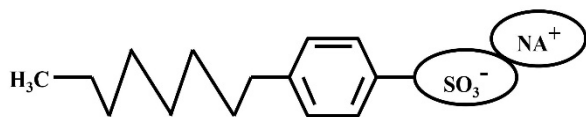


FIGURE 1. Chemical structure of SDBS.

adopted for the preparation of nanofluids were presented. Experimental relative permittivity results of DNFs at different concentrations (Viz., 0.01 wt%, 0.02 wt%, 0.03 wt%, and 0.05 wt%) and varying temperature (Viz., 45 °C, 60 °C, 75 °C and 90 °C) were investigated and compared. A relative permittivity model of composite fluids considering surfactant polarization was proposed. Relative permittivity model results were compared with experimental relative permittivity results. Effect of interfacial zone on improved relative permittivity, AC breakdown voltage and dissipation factor were discussed with the help of experimental results.

II. MATERIALS AND METHODOLOGY

A. MATERIALS

Al₂O₃ NPs were procured from Nanopar Tech, Chandigarh, India. The bulk density of the Al₂O₃ NPs was 0.18 g/mL, with a large surface area, morphology of nearly spherical, and a purity of 99.99%. SDBS was used as a surfactant, since it proved an enhanced stability when being used with Al₂O₃-water nanofluids [26]. It was purchased from Sigma-Aldrich, India, and has the formula of CH₃(CH₂)₁₁C₆H₄SO₃⁻Na⁺ with the structure shown in Fig. 1. Pure, natural and cold pressed cottonseed oil was extracted from cottonseeds purchased from the local Indian market. TBHQ was used as a synthetic antioxidant and it was purchased from Sigma-Aldrich, India. The typical values for physical and chemical properties of cottonseed oil can be found in [27]. Cold pressed cottonseed oil was selected in the current implementation for two reasons. First, cold pressing can retain higher levels of natural antioxidants that may be lost during the refining steps of a commercially available cottonseed oil. Second, cold pressing uses no organic solvent, resulting in chemically contaminant-free oil [28].

B. PREPARATION OF Al₂O₃ NANOFLUIDS

In the current investigation, the cold pressed CSO was used. Preparation of CSO as DBF was already presented in our prior studies [29], [30]. Same procedure was adopted in the current implementation. The cold pressed CSO was introduced with TBHQ (0.02%) and then the solution was heated at 80°C for 90 minutes by magnetic stirrer, thereby removing water and gas contaminants from the oil. Thus, the CSO as DBF became ready for the current implementation. Measured quantities of Al₂O₃ NPs and SDBS surfactant were introduced into CSO DBF and stirred for 20 minutes at room temperature to form DNFs. Al₂O₃ NPs were introduced at 0.01 wt%, 0.02 wt%, 0.03 wt%, and 0.05 wt%, with a 1:1 ratio of SDBS surfactant to Al₂O₃ NPs. The maximum weight fraction of Al₂O₃ NPs was 0.05 wt%, since it has proved stable suspension in natural ester oil [31].

Stable dispersion of DNF samples were attained by Probe Sonicator (Model: PKS-250FM, Anamatrix Instrument Technologies Private Limited, Bangalore, India), with sonication time of 30 minutes at power setting of 50% and pulse setting of 40%. Stability analysis of prepared nanofluids was carried out by visual inspection over a period of four weeks. Nanofluids showed stable dispersion without any aggregates. Fig. 2a includes visual image of Al₂O₃ powder sample and Fig. 2b includes visual images of prepared DNFs (Viz., 0.01 wt%, 0.02 wt%, 0.03 wt%, and 0.05 wt%). Furthermore, to assure the reliability of the obtained results, the dielectric properties were measured shortly after preparation.

III. CHARACTERIZATION OF Al₂O₃ NPs

Fig. 3a and b show transmission electron microscope (TEM, Model: JEM 2100 PLUS, JEOL Ltd., Tokyo, Japan) images of aluminium oxide nanoparticles at different magnification levels. These figures illustrate an aggregate phenomenon of the nanoparticles with a primary particle size of 15–25 nm (±0.5 nm). Selective Area Electron Diffraction (SAED) pattern is shown in Fig. 3c, in which numerous discrete spots line-up and form rings indicating polycrystalline structure. High-resolution TEM image in Fig. 3d indicates multiple planes of different crystals oriented in different directions.

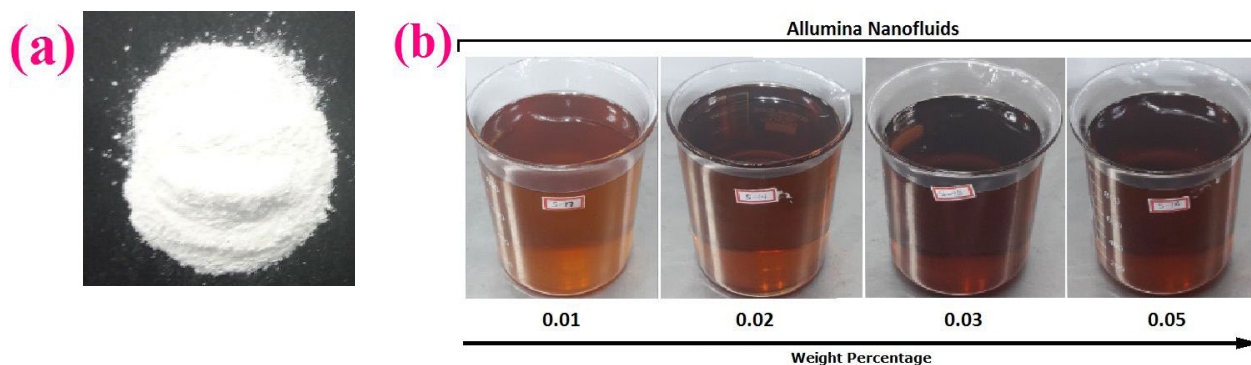


FIGURE 2. Visual images of: (a) Al₂O₃ powder, and (b) Prepared Al₂O₃ nanofluids.

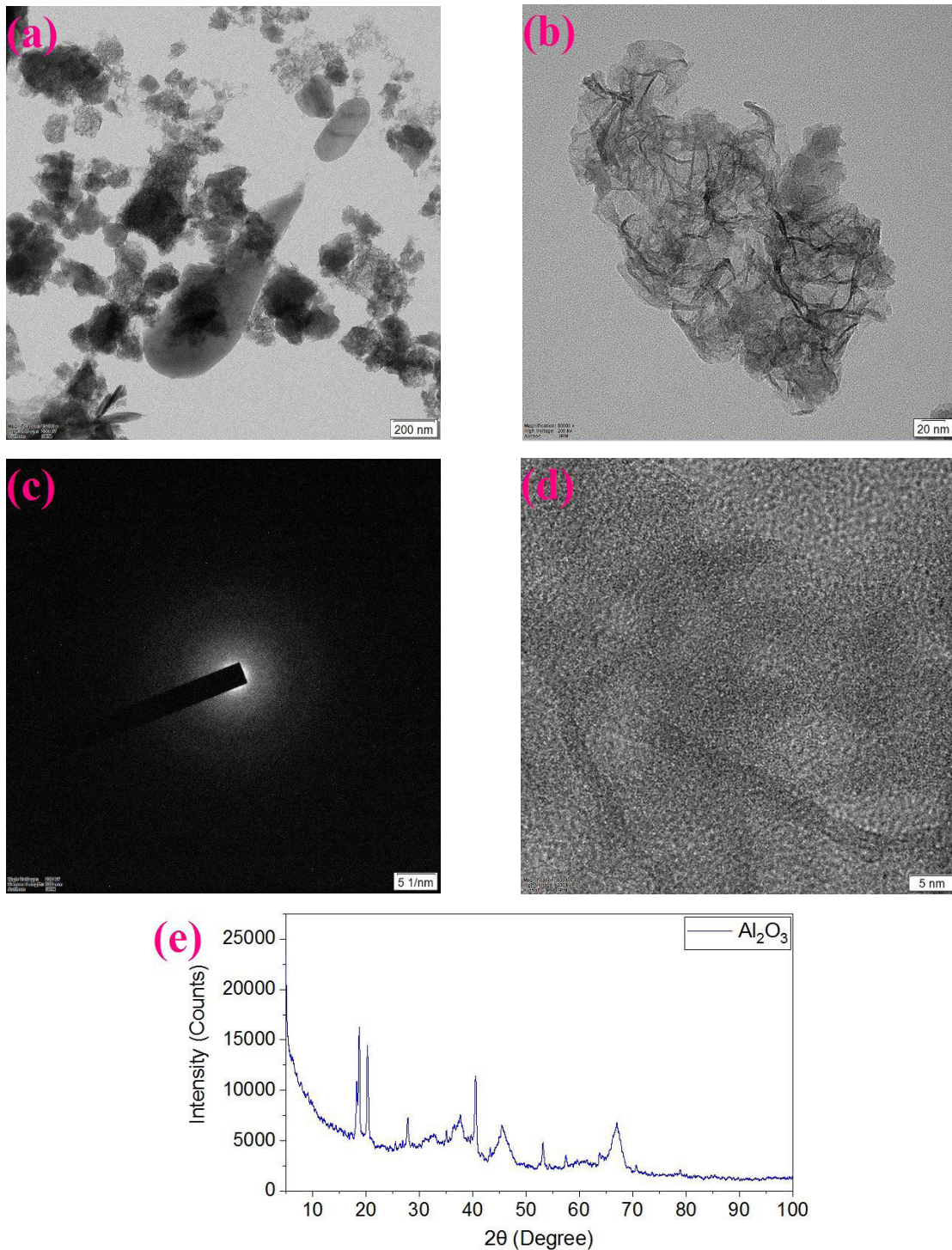


FIGURE 3. Al_2O_3 nanoparticles: (a) TEM image at magnification level 15000 x; (b) TEM image at magnification level 80000 x; (c) SAED pattern; (d) High-resolution TEM image; and (e) XRD pattern.

The X-ray diffraction (XRD, Model: SmartLab SE, Rigaku Corporation, Tokyo, Japan) was obtained for the powder in the 2θ range ($0^\circ - 100^\circ$) and its pattern is displayed in Fig. 3e. The sharp peaks in XRD pattern ensure Al_2O_3 with crystalline structure. From XRD data, the area of crystalline peaks was calculated as 145082 square units and the total area of

all peaks was calculated as 238152 square units, thus the degree of crystallinity was found to be 60.92% referring to the following equation:

$$\text{Crystallinity} = \frac{\text{Area of crystalline peaks}}{\text{Total area}} \times 100 \quad (1)$$

IV. RELATIVE PERMITTIVITY EXPERIMENTAL RESULTS

The relative permittivity or dielectric constant of a dielectric is an AC characteristic and is defined as the increase in capacitance/stored charge, by means of a dielectric medium. The increase in the capacitance/stored charge is attributable to the polarization of the dielectric by the applied electric field [32], as explained in the subsequent section. The relative permittivity of insulating oil in contact with cellulose insulation can affect the local voltage stress distribution. The higher the relative permittivity of oil, the closer its value to that of cellulose insulation and the higher the improvement in electrical stress distribution. Relative permittivity of prepared Al₂O₃ DNFs was measured at various temperatures with ADTR 2K Plus (ELTEL Industries, Bangaluru, India) automatic test set in accordance with the IEC 60247 standard. Relative permittivity was measured at 500 V and 50 Hz AC. Fig. 4 shows the relative permittivity of Al₂O₃ DNFs at different filler levels (Viz., 0.01 wt%, 0.02 wt%, 0.03 wt%, and 0.05 wt%) at four temperature ranges (Viz., 45 °C, 60 °C, 75 °C and 90 °C).

As observed from Fig. 4 the relative permittivity of Al₂O₃ DNFs at a given temperature increases with the increase in the weight percentage from 0.01 wt% to 0.05 wt%. At 45 °C, the relative permittivity increases from 2.98 to 3.87 with the increase in weight percentage from 0.01 wt% to 0.05 wt%. The relative permittivity of Al₂O₃ DNFs at a given weight percentage is also a function of the temperature, where it decreases with the increase in temperature. The temperature dependency of relative permittivity has similar trend to that obtained in literature [29], [33], [34]. The relative permittivity at 45 °C was measured as 2.98, 3.21, 3.44, and 3.87 for 0.01 wt%, 0.02 wt%, 0.03 wt%, and 0.05 wt% DNFs, respectively. Whereas, at 90 °C for the same DNFs, it was measured as 2.83, 2.99, 3.21, and 3.67, respectively.

The increase in the relative permittivity of DNFs with the increase in weight percentage of Al₂O₃ NPs can be attributed to internal polarization of NPs and orientational polarization of charged NPs referring to the Clausius-Mossotti equation as follows:

$$\frac{\epsilon_{NF} - 1}{\epsilon_{NF} + 2} = \frac{1}{3\epsilon_0} [N_1\alpha_1 + N_2\alpha_2 + N_2\alpha_3] \quad (2)$$

As both internal polarization of NPs and orientational polarization of charged NPs increase with the increase in the number of nanoparticles, so the relative permittivity increases against the weight percentage of NPs. The decreasing trend in relative permittivity with the increase in temperature at each weight percentage of Al₂O₃ NPs was observed and was justified with the proposed model in the next section.

V. RELATIVE PERMITTIVITY MODEL WITH SURFACTANT CONSIDERATION

DNFs under the influence of an external electrical field, both base oil molecules and NPs will get polarized. Surface polarization charges (positive and negative) are accumulated

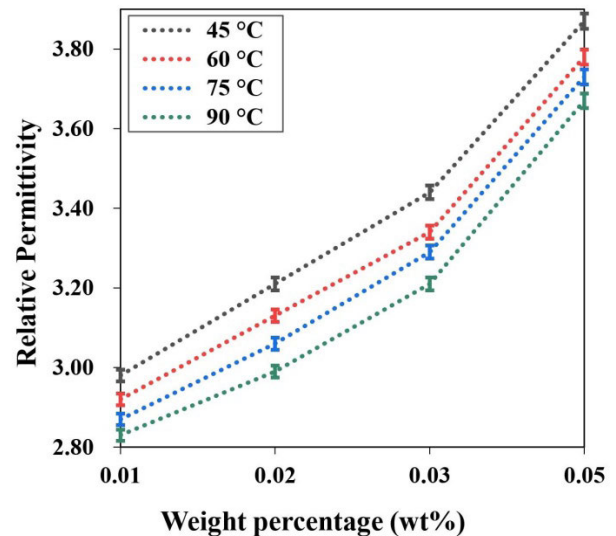


FIGURE 4. Experimental relative permittivity results of Al₂O₃ nanofluids at varying temperatures and weight percentages.

on either sides of the nanoparticle, resulting in the particle being charged. There are three types of polarization in DNFs that determine the relative permittivity of DNF, as well as the local voltage stress distribution under external electrical field. Namely, polarization of base fluid molecules, internal polarization of NPs, and orientational polarization of charged NPs. Clausius-Mossotti equation expresses the relative permittivity of DNFs in terms of polarization as depicted above in equation (2).

Equation (2) can be expressed as follows:

$$\frac{\epsilon_{NF} - 1}{\epsilon_{NF} + 2} = \frac{N_1\alpha_1}{3\epsilon_0} + \frac{N_2\alpha_2}{3\epsilon_0} + \frac{N_2\alpha_3}{3\epsilon_0} \quad (3)$$

For the pure dielectric fluid (without nanoparticle), equation (3) is simplified as follows:

$$\frac{\epsilon_1 - 1}{\epsilon_1 + 2} = \frac{N_1\alpha_1}{3\epsilon_0} \quad (4)$$

By definition, polarizability of NPs can be expressed as follows [32]:

$$\alpha_2 = \epsilon_0 (\epsilon_2 - 1) V \quad (5)$$

Assume nanoparticles are spherical, the number of nanoparticles per unit volume can be calculated as follows:

$$N_2 = \frac{\varphi_{NP}}{V} = \frac{\varphi_{NP}}{\frac{4}{3}\Pi a^3} = \frac{3\varphi_{NP}}{4\Pi a^3} \quad (6)$$

From equations (5) and (6), the following equation can be obtained:

$$\frac{N_2\alpha_2}{\epsilon_0} = \varphi_{NP} (\epsilon_2 - 1) \quad (7)$$

The charge density of NPs surface polarization excluding surfactant polarization can be expressed as follows [14]:

$$\sigma_1 = \epsilon_0 E_0 \left(\frac{\epsilon_2 - \epsilon_1}{\epsilon_2 + 2\epsilon_1} \right) \sin \theta \cos \varphi \quad (8)$$

To consider surfactant polarization under an external electric field, the polarization model in Fig. 5 is proposed. The polarization charges, including positive and negative charges, are accumulated on both sides of the spherical NP and opposite to the external electrical field as shown in Fig. 5a. As a result, surfactant encapsulates NP with opposite polarization charges on the inner side to neutralize net charges of NP. While on the outer side, same amount of opposite charges gets distributed. Fig. 5b shows the same polarization NP model with illustrating all dimensions of NP and surfactant as well as the magnitude of polarization charges.

Considering surfactant polarization, surface charge density on NP can be expressed as follows [16]:

$$\sigma_2 = \epsilon_0 E_0 \left[\left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) - \left(\frac{ad + d^2}{a} \right) \left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) \right] \times \sin \theta \cos \varphi \quad (9)$$

Referring to Fig. 6 and considering surfactant polarization, the total positive charge (Q_+) on NPs can be calculated as follows:

$$dQ = \sigma_2 a \, d\theta \, a \, \sin \theta \, d\varphi \quad (10)$$

$$Q_+ = 4 \int_0^{\pi/2} \int_0^{\pi/2} \sigma_2 a^2 \sin \theta \, d\theta \, d\varphi \quad (11)$$

$$Q_+ = \Pi a^2 \epsilon_0 E_0 \left[\left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) - \left(\frac{ad + d^2}{a} \right) \left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) \right] \quad (12)$$

According to the dielectric concept of polar molecules, the electric dipole moment of NPs molecules can be expressed as follows:

$$p = 2a \, Q_+ \quad (13)$$

Dipolar orientational polarizability α_3 per molecule can be defined as follows:

$$\alpha_3 = \frac{p^2}{3kT} = \frac{4a^2 Q_+^2}{3kT} \quad (14)$$

Substituting the value of Q_+ in equation (14), the following equation can be obtained:

$$\alpha_3 = \frac{4a^6}{3kT} \Pi^2 \epsilon_0^2 E_0^2 \times \left[\left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) - \left(\frac{ad + d^2}{a} \right) \left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) \right]^2 \quad (15)$$

Thus, substituting fourth term of equation (6) and (15), the third term in equation (2) can be expressed as follows:

$$\frac{N_2 \alpha_3}{\epsilon_0} = \frac{1}{kT} \Pi \epsilon_0 a^3 \varphi_{NP} E_0^2 \times \left[\left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) - \left(\frac{ad + d^2}{a} \right) \left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) \right]^2 \quad (16)$$

Substituting for equations (4), (7), and (16) into equation (2), the polarization model of DNF considering

TABLE 1. Parameters considered in calculating the relative permittivity of nanofluids according to the proposed model.

No.	Parameter	Value
1	2a	20 nm
2	D	1.5 nm
3	ϵ_1	2.941 at 45 °C, 2.879 at 60 °C, 2.84 at 75 °C and 2.799 at 90 °C
4	ϵ_2	7.8
5	ϵ_0	$8.854187817 \times 10^{-12} \text{ F.m}^{-1}$
6	φ_{NP}	0.01 wt%, 0.02 wt%, 0.03 wt%, and 0.05 wt%
7	k	$1.380649 \times 10^{-23} \text{ J.K}^{-1}$
8	T	318.15 K, 333.15 K, 348.15 K, and 363.15 K.

surfactant polarization can be obtained as follows:

$$\frac{\epsilon_{NF} - 1}{\epsilon_{NF} + 2} = \frac{\epsilon_1 - 1}{\epsilon_1 + 2} + \frac{\varphi_{NP} (\epsilon_2 - 1)}{3} + \frac{1}{3kT} \Pi \epsilon_0 a^3 \varphi_{NP} E_0^2 \times \left[\left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) - \left(\frac{ad + d^2}{a} \right) \left(\frac{\epsilon_2 - \epsilon_1}{2\epsilon_1 + \epsilon_2} \right) \right]^2 \quad (17)$$

VI. COMPARISON OF THE PROPOSED MODEL WITH EXPERIMENTAL RELATIVE PERMITTIVITY RESULTS

In this section, the proposed model in the previous section is compared to the experimental results. As indicated from TEM images in Section III, primary particle size of Al_2O_3 was in the range from 15 to 25 nm. To calculate relative permittivity of DNFs according to the proposed model, the average diameter of NPs is considered 20 nm, the thickness of surfactant is considered 1.5 nm [16], and the external electric field applied is 500 V. Other parameters are depicted in Table 1.

Comparison between results of the proposed model and experimental results at 45 °C, 60 °C, 75 °C, and 90 °C are depicted in Fig. 7. The values of relative permittivity calculated are in good agreement with that obtained experimentally. At a given temperature and weight percentage, there is a slight difference between the results obtained from the model and the experimental results. The percentage difference between the experimental results and the model results are depicted in Table 2, lowest difference is 3.48% and highest difference is 6.16%. Accordingly, the developed model will enable to accurately calculate the gross relative permittivity of nanofluids before actual development.

VII. EXPERIMENTAL RESULTS OF TAN δ

The relative magnitude of dielectric loss with respect to dielectric constant is defined as $\tan \delta$. It is also called the loss factor (or loss tangent). The $\tan \delta$ of prepared Al_2O_3 DNFs was measured at various temperatures with ADTR 2K Plus (ELTEL Industries, Bangaluru, India) automatic test set in accordance with the IEC 60247 standard. $\tan \delta$ is an AC characteristic and was measured at 500 V and 50 Hz AC. Fig. 8 shows the $\tan \delta$ of Al_2O_3 DNFs at different filler

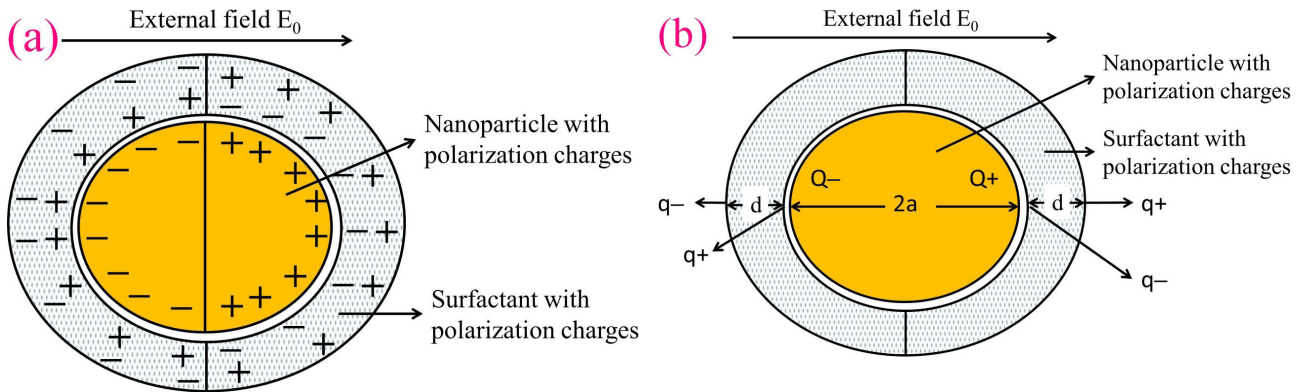


FIGURE 5. Polarization model of nanoparticle post electrical field application on nanofluid (a) Polarization charges; and (b) Dimensions and charge magnitude of nanoparticle and surfactant.

TABLE 2. Experimental and model result percentage difference.

Temperature → wt% ↓	45 °C	60 °C	75 °C	90 °C
0.01	4.87 %	4.99 %	5.36 %	5.28 %
0.02	3.88 %	4.39 %	5.29 %	6.16 %
0.03	3.48 %	4.36 %	4.45 %	5.44 %
0.05	5.03 %	5.33 %	5.17 %	5.22 %

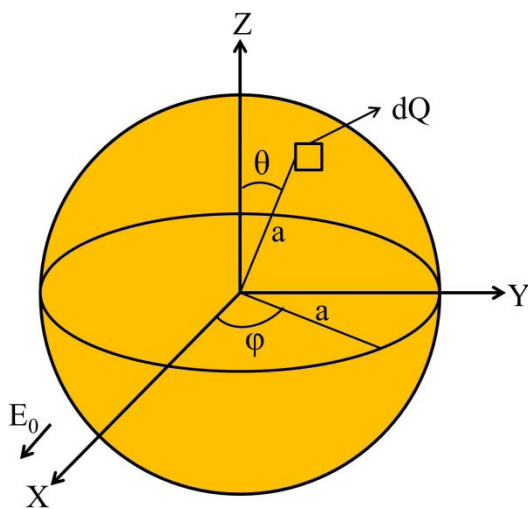


FIGURE 6. Surface polarization charges on nanoparticle post electrical field application on nanofluid.

levels (Viz., 0.01 wt%, 0.02 wt%, 0.03 wt%, and 0.05 wt%) and at four temperature ranges (Viz., 45 °C, 60 °C, 75 °C and 90 °C).

As observed from Fig. 8, the $\tan \delta$ of Al_2O_3 DNFs at a given temperature decreases with the increase in the weight percentage from 0.01 wt% to 0.05 wt%. At 45 °C, $\tan \delta$ decreases from 0.043 to 0.016 with the increase in weight percentage from 0.01 wt% to 0.05 wt%. The $\tan \delta$ of Al_2O_3 DNFs is also a function of temperature at a given weight percentage. In fact, $\tan \delta$ increases with the increase in tem-

perature which is in line with the literature [35]. At 0.01 wt%, the $\tan \delta$ increases from 0.043 to 0.1656 with the increase in temperature from 45 °C to 90 °C, while at 0.05 wt%, the $\tan \delta$ increases from 0.016 to 0.0716 with the same increase in temperature.

The decrease in $\tan \delta$ of DNFs with the increase in weight percentage of Al_2O_3 NPs is attributed to the increase of dielectric constant and the decrease of dielectric loss in the denominator and numerator of $\tan \delta$, respectively. For dielectric constant, it increases with the increase in weight percentage due to the increase in number of Al_2O_3 NPs as discussed in the earlier section. On the other hand, the charge trapping provided by nanoparticles and their interfaces decreases the dielectric loss [29]. Thus, the total effect is the decrease of $\tan \delta$ against the weight percentage. This decrease in $\tan \delta$ against the weight percentage is more pronounced at 90 °C than that at 45 °C. This trend can be attributed to the effect of temperature on the number of charge carriers into the fluid and Brownian motion of nanoparticles. Increasing the temperature causes an increase in the number of charge carriers, thereby increasing the value of dielectric loss and $\tan \delta$. On the other hand, increasing the temperature causes a subsequent increase in Brownian motion of nanoparticles enabling to trap more charge carriers, thereby decreasing the value of dielectric loss and $\tan \delta$. With increasing the weight percentage of nanoparticles, the temperature effect on Brownian motion increases, while the temperature effect on the number of charge carriers remains constant, causing the decrease in $\tan \delta$ against the weight percentage to be more pronounced at 90 °C.

VIII. AC BREAKDOWN VOLTAGE EXPERIMENTAL RESULTS

AC breakdown voltage measures the ability of transformer oil to withstand high voltages during real-time application. AC BDV of DNFs was measured as per IEC 60156, whose specifications include sphere to sphere electrode, 2.5 mm gap and 2 kV/s voltage ramp rate. The mean value of AC BDV for the DBF at room temperature and pressure was measured as 37.6 kV.

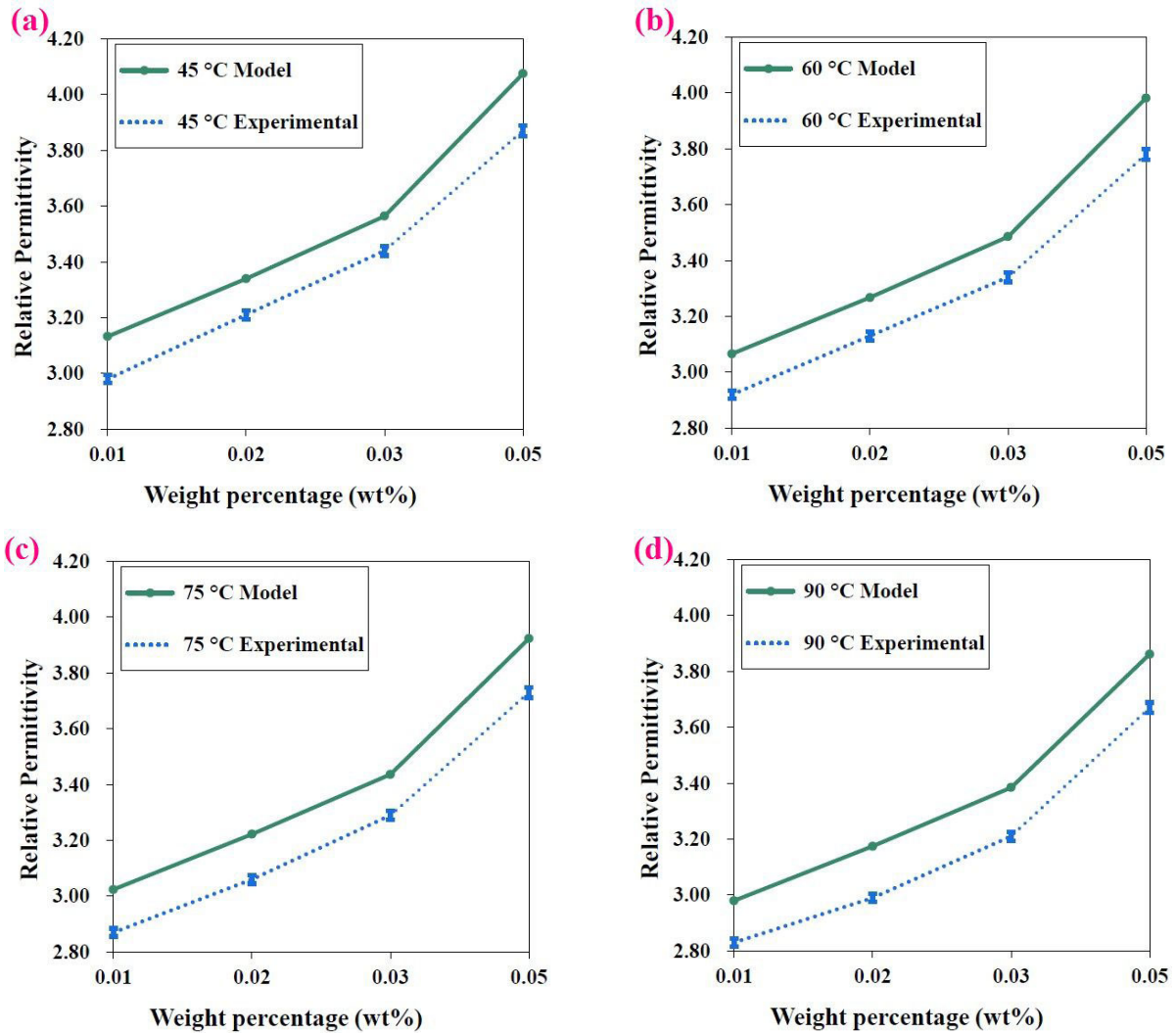


FIGURE 7. Relative permittivity of nanofluids according to the proposed model at various temperatures: (a) 45 °C, (b) 60 °C, (c) 75 °C and (d) 90 °C.

The mean AC BDV of Al₂O₃ DNFs is incremental with the weight percentage of Al₂O₃ NPs (Viz., 0.01 wt%, 0.02 wt%, 0.03 wt% and 0.05 wt%) as depicted in Fig. 9. This increment is attributed to the increase in trapping depth provided by the increase in weight percentage of Al₂O₃ NPs. Maximum AC BDV was measured at 0.05 wt% of Al₂O₃ DNF and it was measured as 46.2 kV corresponding to a percentage enhancement of about 23%. Mean, standard deviation (SD), and percentage enhancement in AC BDV of DNFs is listed in Table 3.

IX. DISCUSSION
A. INTERFACIAL ZONE

It is known that Al₂O₃ nanoparticles absorbs H⁺ or OH⁻ species by water absorbance from moisture existing in the oil. This depends upon acid-base equilibrium which is determined by the isoelectric point of nanoparticles and pH of base oil. The isoelectric point is a point at which nanoparticles

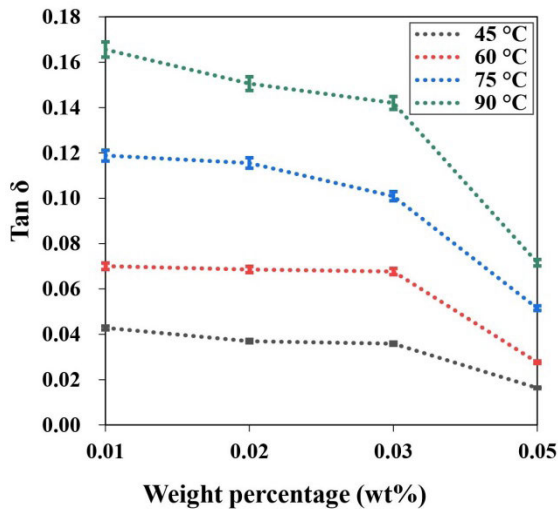
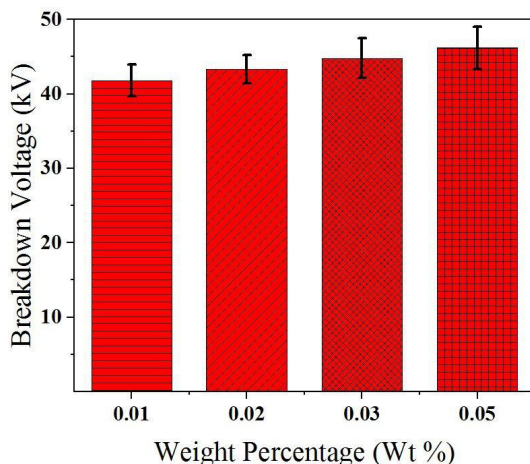
TABLE 3. AC BDV test results of Al₂O₃ DNFs.

Sample (wt %)	AC BDV		
	Mean (kV)	SD	Enh. (%)
0.01	41.8	2.15	11.17
0.02	43.3	1.91	15.15
0.03	44.8	2.71	19.14
0.05	46.2	2.85	22.87

carriers no net charges on their surface. Since pH value of cottonseed oil is around 7 [36] and the isoelectric point of Al₂O₃ is 9.4 [37], Al₂O₃ nanoparticles absorb H⁺ species on O atoms and get positive charges on their surface. Accordingly, the usage of an anionic surfactant like SDBS facilitates its adsorption on the surface of nanoparticles leaving behind

TABLE 4. Relative permittivity due to orientational polarization.

wt% ↓	45 °C ↓		60 °C ↓		75 °C ↓		90 °C ↓	
	Without	With	Without	With	Without	With	Without	With
0.01	6.65×10^{-13}	1.49×10^{-14}	6.64×10^{-13}	3.49×10^{-13}	6.52×10^{-13}	3.43×10^{-13}	6.44×10^{-13}	3.38×10^{-13}
0.02	1.33×10^{-12}	6.99×10^{-13}	3.32×10^{-12}	6.98×10^{-13}	1.30×10^{-12}	6.86×10^{-13}	1.29×10^{-12}	6.77×10^{-13}
0.03	2.00×10^{-12}	1.05×10^{-12}	1.99×10^{-12}	1.05×10^{-12}	1.96×10^{-12}	1.03×10^{-12}	1.93×10^{-12}	1.02×10^{-12}
0.05	3.33×10^{-12}	1.75×10^{-12}	3.32×10^{-13}	1.74×10^{-12}	3.26×10^{-12}	1.72×10^{-12}	3.22×10^{-12}	1.69×10^{-12}

**FIGURE 8.** Tan δ of Al_2O_3 nanofluids at varying temperatures and weight percentages.**FIGURE 9.** AC BDV test results of Al_2O_3 DNFs.

NaOH as shown in Fig. 10. This results in a sufficient repulsion force between nanoparticles and an enhancement in the dispersion of nanoparticles with positive impacts on thermal and dielectric properties.

B. IMPACT OF INTERFACIAL ZONE ON DIELECTRIC PROPERTIES

Since, surfactants' heads are adsorbed on the surface of nanoparticles and their tails are directed towards oil,

surfactants' molecules become aligned in parallel to each other and perpendicular to the surface of nanoparticles. The intermolecular forces keep the molecules staying apart from each other. As a result, oil chains in this region are forced to be aligned between surfactants' molecules creating two different layers [38] as shown in Fig. 11. The first layer is the aligned layer where oil chains are arranged perpendicular to nanoparticle surface and parallel to surfactants' molecules. The second layer is the affected layer, which has less aligned oil chains but is affected by the first layer. In case of nanofluids without surfactant there will be only the affected layer due to confinement of oil chains in the vicinity of nanoparticles at the interfacial zone.

For nanofluids with SDBS surfactant, the obtained dielectric properties are referred to interfacial zone. As mentioned above the interfacial zone is composed of two different layers. The aligned layer has a rigid structure that is difficult to polarize. Due to this rigid structure, the total amount of polarization decreases resulting in a decrease of dielectric constant with the addition of surfactant. This rigid structure also makes it easy for electrons to lose their energy resulting in an increase in breakdown strength and a decrease in dielectric losses.

C. COMPARISON OF PROPOSED MODEL FOR SURFACTANT TREATED AND NON-TREATED NANOPARTICLES

Referring to proposed model, the first and second terms on the right hand side of equation (17) represent relative permittivity contribution by polarization of base oil molecules and inner polarization of nanoparticles, respectively, while the third term represents relative permittivity contribution by orientational polarization. Therefore, the first and second terms remain the same for nanoparticles either treated with surfactant or non-treated without surfactant. Whereas, the third term changes for nanoparticles treated with and without surfactant. Table 4 shows relative permittivity contribution due to orientational polarization with and without surfactant at different filler levels (Viz., 0.01 wt%, 0.02 wt%, 0.03 wt%, and 0.05 wt%) and at four temperature ranges (Viz., 45 °C, 60 °C, 75 °C and 90 °C). It is worthy mentioned that the relative permittivity contribution without surfactant calculated by the present proposed model matches well with the model proposed earlier in [14]. This validates the proposed model. However, the current research presents

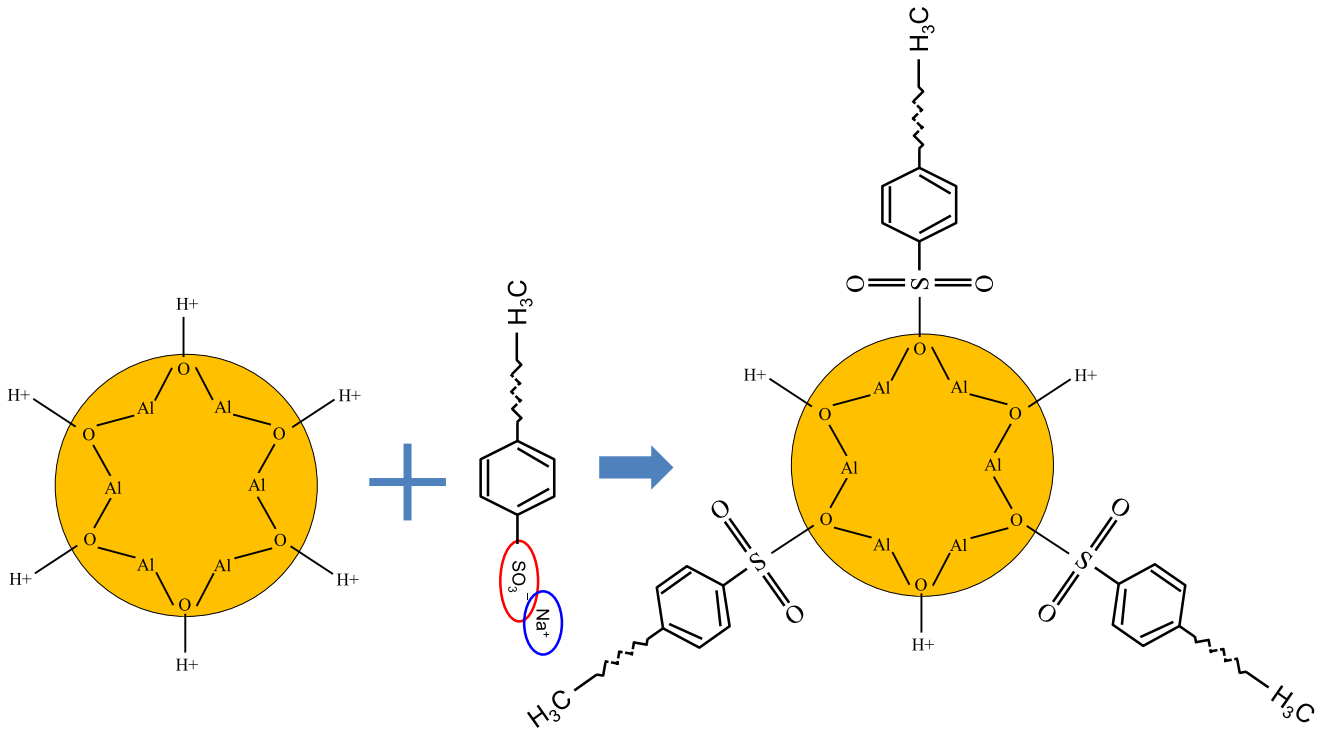


FIGURE 10. The adsorption of SDBS surfactant on the surface of Al_2O_3 nanoparticles.

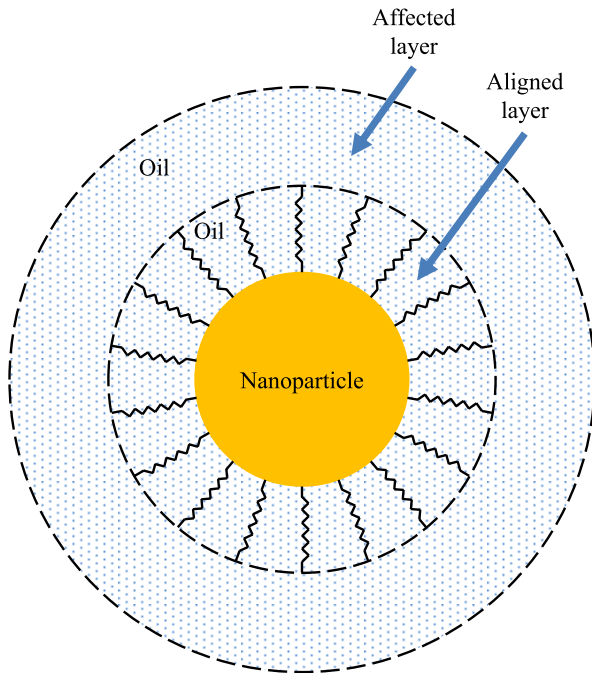


FIGURE 11. The structure of interfacial zone around nanoparticles in cottonseed oil.

an extended part with consideration of surfactant effect. It is clear that there is a decrement in the relative permittivity due to orientational polarization after surfactant treatment compared to without surfactant treatment at a given temperature and filler levels of nanoparticles. As discussed earlier,

this is due to the effect of aligned layer that is difficult to polarize.

X. CONCLUSION

Effect of interfacial zone on dielectric properties of cottonseed oil based Al_2O_3 DNFs was investigated. Detailed characterization of Al_2O_3 nanoparticles and methodology adopted for the preparation of DNFs were presented. A relative permittivity model of DNFs considering surfactant polarization was proposed. Experimental relative permittivity results of DNFs at varying temperature were investigated. Relative permittivities of proposed model were compared with experimental results. Effect of interfacial zone on dissipation factor and AC BDV were also discussed with the help of experimental results and corresponding physical mechanisms. The conclusions made are as follows:

1. The relative permittivity of Al_2O_3 DNFs increased with the increase in the weight percentage of NPs. This was explained considering internal polarization of NPs and orientational polarization of charged NPs.
2. The relative permittivity of Al_2O_3 DNFs decreased with the increase in temperature, which is a similar trend to that obtained in literature.
3. From the proposed model, it was found that surfactant contributes for orientational polarization of Al_2O_3 nanoparticles in Clausius-Mossotti equation for the relative permittivity of DNFs.
4. From the proposed model, the relative permittivity of DNFs was found independent of the diameter of nanoparticles (from the range $2a = 15nm$ to $25nm$) and

the thickness of surfactant (from the range $d = 1.5\text{nm}$ to 3.0nm), but instead it was highly dependent on the number of nanoparticles per unit volume, thus indirectly depends on the dispersion state.

5. Relative permittivities of proposed model were almost in line with experimental results, at a given weight percentage of Al_2O_3 NPs and at a given temperature. The highest percentage difference between experimental and model results was 6.16%.
6. The dissipation factor of DNFs decreased with the increase in weight percentage of Al_2O_3 NPs due to the increase of relative permittivity that clarified from the proposed model and the decrease of dielectric loss that provided by charge trapping of nanoparticles and their interfaces.
7. In the current study, concentrations up to 0.05 wt% were found to be below or equal to the critical concentration. Therefore, the mean AC BDV of surfactant driven nanofluid was incremental with the weight percentage of Al_2O_3 NPs due to the increase in trapping depth. Maximum AC BDV was measured as 46.2 kV corresponding to a percentage enhancement of about 23%.

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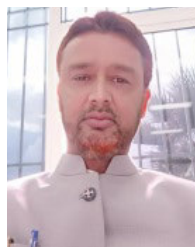
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