

# UNIVERSITI PUTRA MALAYSIA

DIELECTRIC AND THERMAL PROPERTIES OF NATURAL ESTER GREEN NANOFLUIDS BASED ON COTTONSEED OIL FOR TRANSFORMER APPLICATIONS

**RIJAVAN FARADE** 

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## DIELECTRIC AND THERMAL PROPERTIES OF NATURAL ESTER GREEN NANOFLUIDS BASED ON COTTONSEED OIL FOR TRANSFORMER APPLICATIONS



Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy

December 2021

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## DEDICATION

This thesis is gratefully dedicated to:

My beloved Parents for their love, patience and understanding

&

My beloved wife, daughters, brother and in-law.



Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

### DIELECTRIC AND THERMAL PROPERTIES OF NATURAL ESTER GREEN NANOFLUIDS BASED ON COTTONSEED OIL FOR TRANSFORMER APPLICATIONS

By

#### **RIJAVAN FARADE**

December 2021

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Vegetable oils have emerged as insulating fluids in transformer applications and as a prominent and effective alternative for traditional dielectric fluids. However, most vegetable oils are edible and their application is limited on a large scale. In the present work, a non-edible vegetable oil is developed as an insulating fluid. The developed oil is oxidation-inhibited cottonseed oil (CSO). Tertiary butylhydroquinone (TBHQ) was used as antioxidant. The concept of nanofluids was used to overcome the limited dielectric and thermal properties of base CSO. The used types of nanoparticles (NPs) were hexagonal boron nitride (h-BN) with 0.01 - 0.1 wt%, graphene oxide (GO) with 0.01 - 0.05 wt%, graphene with 0.0015 - 0.01 wt%, and Al<sub>2</sub>O<sub>3</sub> NP with 0.01 - 0.05 wt%. Characterization of these NPs, methodology adopted for the preparation of nanofluids, and their stability into the prepared nanofluids were presented. GO was treated with sodium dodecyl sulfonate (SDS), graphene and  $Al_2O_3$  with sodium dodecylbenzene sulfonate (SDBS) to obtain stable suspensions. The prepared nanofluids were tested for dielectric and thermal properties. The dielectric properties include breakdown voltages at room temperature and pressure; dielectric constant, dissipation factor, and resistivity at at 45 °C, 60 °C, 75 °C, and 90 °C; while thermal properties include thermal conductivity at 35 °C, 45 °C, 55 °C, and 65 °C; followed by thermal response and thermogram analysis. Few researchers investigated stability of dielectric nanofluids for use in transformers and adopted less effective qualitative visual inspection method. Also, relative permittivity models proposed earlier did not consider the interfacial zone created by surfactant. Therefore, three main objectives were aimed in this research; to develop non-edible CSO based nanofluids; to enhance dispersion stability, dielectric and thermal properties of CSO based nanofluids; and to design a relative permittivity model of nanofluids created by interfacial zone considering surfactant polarization. To attain the first objective, dielectric and thermal properties of CSO based h-BN and GO nanofluids were comprehensively evaluated. To attain the second objective, CSO based graphene nanofluids prepared with different sonication times were investigated for long-term stability through effective Ultraviolet-visible (UV-Vis) spectroscopy. Also, sonication time impact on dielectric and thermal properties were investigated. To attain the third objective, a polarization model of nanofluids was proposed to calculate relative permittivity considering surfactant effect. For the first objective, all the obtained results validated the superiority of CSO based h-BN and GO nanofluids as a potential candidates for power equipment. For the second objective, the presented results showed the relation between dispersion stability and thermo-dielectric properties, thereby contributed to long-term stability of CSO based graphene nanofluids. For the third objective, the results were discussed considering the structure of interfacial zone of CSO based Al<sub>2</sub>O<sub>3</sub> nanofluids and the relative permittivity calculated from the model was almost in line with experimental results. In addition, for all the obtained results of the proposed nanofluids, corresponding physical mechanisms were discussed and clarified. Thus, dielectric and thermal properties investigation of proposed CSO based nanofluids open up a great opportunity in natural ester insulating fluid applications.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

### SIFAT DIELEKTRIK DAN TERMA ESTER SEMULAJADI CECAIR NANO HIJAU BERASASKAN MINYAK BIJI KAPAS UNTUK APLIKASI TRANSFORMER

Oleh

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Minyak sayuran telah muncul sebagai cecair penebat dalam aplikasi transformer dan sebagai alternatif yang menonjol dan berkesan untuk cecair dielektrik tradisional. Walau bagaimanapun, kebanyakan minyak sayuran boleh dimakan dan penggunaannya terhad pada skala besar. Dalam kerja ini, minyak sayuran tidak boleh dimakan dibangunkan sebagai cecair penebat. Minyak yang dibangunkan ialah minyak biji kapas (CSO) yang pengoksidaanny-terhalang. Butylhydroquinone tertier (TBHQ) digunakan sebagai antioksidan. Konsep cecair nano digunakan untuk mengatasi sifat dielektrik dan terma yang terhad bagi CSO asas. Empat jenis nanopartikel (NP) yang digunakan ialah hexagonal boron nitride (h-BN) dengan 0.01 - 0.1 wt%, graphene oxide (GO) dengan 0.01 - 0.05 wt%, graphene dengan 0.0015 - 0.01 wt%, dan Al<sub>2</sub>O<sub>3</sub> NP dengan 0.01 - 0.05wt%. Pencirian NP ini, metodologi yang digunakan untuk penyediaan cecair nano, dan kestabilan mereka ke dalam cecair nano yang disediakan telah dibentangkan. GO dirawat dengan sodium dodecyl sulfonate (SDS), graphene dan Al<sub>2</sub>O<sub>3</sub> dengan sodium dodecylbenzene sulfonate (SDBS) untuk mendapatkan suspensi yang stabil. Bendalir nano yang disediakan telah diuji untuk sifat dielektrik dan terma. Pengkajian sifat dielektrik termasuk voltan kerosakan pada suhu dan tekanan bilik; pemalar dielektrik, faktor pelesapan dan kerintangan pada 45 °C, 60 °C, 75 °C dan 90 °C; manakala sifat terma termasuk kekonduksian terma pada 35 °C, 45 °C, 55 °C dan 65 °C; diikuti dengan tindak balas terma dan analisis termogram. Beberapa penyelidik menyiasat kestabilan cecair nano dielektrik untuk digunakan dalam transformer dan menggunakan kaedah pemeriksaan visual kualitatif yang kurang berkesan. Juga, model kebolehtelapan relatif yang dicadangkan sebelum ini tidak mengambil kira zon antara muka yang dicipta oleh surfaktan. Oleh itu, tiga objektif utama telah disasarkan dalam penyelidikan ini; untuk membangunkan cecair nano berasaskan CSO yang tidak boleh dimakan; untuk meningkatkan kestabilan serakan, sifat dielektrik dan terma bagi cecair nano berasaskan CSO; dan untuk mereka bentuk model ketelusan relatif bagi cecair nano yang dicipta oleh zon antara muka dengan mengambil kira polarisasi surfaktan. Untuk mencapai objektif pertama, sifat dielektrik dan terma bagi cecair nano h-BN dan GO berasaskan CSO dinilai secara menyeluruh. Untuk mencapai objektif kedua, cecair nano graphene berasaskan CSO yang disediakan dengan masa sonikasi berbeza telah dikaji untuk kestabilan jangka panjang melalui spektroskopi Ultraviolet-visible (UV-Vis) yang berkesan. Juga, kesan masa sonikasi ke atas sifat dielektrik dan terma telah disiasat. Untuk mencapai objektif ketiga, model polarisasi cecair nano telah dicadangkan untuk mengira kebolehtelapan relatif mempertimbangkan kesan surfaktan. Untuk objektif pertama, semua keputusan yang diperolehi mengesahkan keunggulan h-BN dan GO berasaskan CSO cecair nano sebagai calon berpotensi untuk peralatan kuasa. Bagi objektif kedua, keputusan yang dibentangkan menunjukkan hubungan antara kestabilan serakan dan sifat termo-dielektrik, dengan itu menyumbang kepada kestabilan jangka panjang cecair nano graphene berasaskan CSO. Untuk objektif ketiga, keputusan telah dibincangkan dengan mengambil kira struktur zon antara muka CSO berasaskan Al<sub>2</sub>O<sub>3</sub> cecair nano dan kebolehtelapan relatif yang dikira daripada model adalah hampir selari dengan keputusan eksperimen. Di samping itu, untuk semua keputusan yang diperolehi daripada cecair nano yang dicadangkan, mekanisme fizikal yang sepadan telah dibincangkan dan dijelaskan. Oleh itu, penyiasatan sifat dielektrik dan terma bagi cecair nano berasaskan CSO yang dicadangkan membuka peluang besar dalam aplikasi cecair penebat ester semula jadi.

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**Rijavan Farade** February 2022 This thesis was submitted to the Senate of the Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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## **Declaration by Members of Supervisory Committee**

This is to confirm that:

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- the research conducted and the writing of this thesis was under our supervision;
- supervision responsibilities as stated in the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) are adhered to.

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## LIST OF ABBREVIATIONS

### Nomenclature

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Coulomb constant [8.9875517923×10 <sup>9</sup> kg·m <sup>3</sup> ·s <sup>-2</sup> ·C <sup>-2</sup> ]
Radius of spherical nanoparticle [m]
Absorbance in y intercept [Au]
Breakdown voltage [k.V]
Mode specific heat of branch <i>i</i> . [J.kg <sup>-1</sup> .K <sup>-1</sup> ]
Specific heat capacity [J.kg- <sup>1</sup> .K <sup>-1</sup> ]
Cottonseed oil
Surfactant thickness [m]
Dielectric base fluid
Dielectric nanofluid
Nanoparticle diameter [m]
Charge on nanoparticle's small volume [C]
External electric field applied in the direction of x-axis [V]
Electrical double layer
Energy-Dispersive X-Ray
Enhancement
Hexagonal boron nitride
Branch index
Charging current [A]
Effective current [A]
Boltzmann constant [1.3806505e <sup>-23</sup> J.K <sup>-1</sup> ]
Thermal conductivity due to Brownian motion [W.m <sup>-1</sup> .K <sup>-1</sup> ]

	K <sub>EDL</sub>	Thermal conductivity term according to EDL [W.m <sup>-1</sup> .K <sup>-1</sup> ]
	$K_p$	Thermal conductivity due to phonon transport [W.m <sup>-1</sup> .K <sup>-1</sup> ]
	L <sub>NP</sub>	Mean free path of nanoparticles [m]
	L <sub>oil</sub>	Mean free path of oil molecules [m]
	m	Coefficient of molar extinction [M <sup>-1</sup> .cm <sup>-1</sup> ]
	$M_{np}$	Nanoparticle mass [kg]
	Ν	Concentration of nanoparticles
	n	Number of phonon polarization branches
	N1, N2 & N3	No. of molecules of oil, nanoparticle, and oil at nanoparticle/oil interface
	NP	Nanoparticle
	р	Electric dipole moment of nanoparticles [C·m]
	q	Surface charge of nanoparticles [C]
	Q+	Total positive charge on NPs [C]
	RTP	Room temperature and pressure
	SAED	Selected area electron diffraction
	SD	Standard deviation
	SDBS	Sodium dodecylbenzene sulfonate
	SDS	Sodium dodecyl sulfate
	SEM	Scanning Electron Microscope
	Т	Temperature [K]
	TBHQ	Tertiary butylhydroquinone
	TEM	Transmission Electron Microscope
	UV-Vis	Ultraviolet-visible
	V	Volume [m <sup>3</sup> ]

$v_i$	Phonon group velocity of branch $i$ . [m.s <sup>-1</sup> ]	
wt%	Weight percentage	
x	Concentration [mol.m <sup>-3</sup> ]	
XRD	Powder X-ray diffraction	
У	Absorbance [Au]	

## Greeks

μ	Viscosity of medium [N.s.m <sup>-2</sup> ]
$\alpha_1$	Polarizability of base oil molecules
α <sub>2</sub>	Inner polarizability of NPs
α <sub>3</sub>	Chapter 4, polarizability of oil at nanoparticle/oil interface; Chapter 7, orientational polarizability of charged NPs
3	Relative permittivity
ε'	Dielectric constant
۳"	Loss factor
ε <sub>0</sub>	Permittivity of space [F.m <sup>-1</sup> ]
ει	Relative permittivity of base fluid
ε2	Relative permittivity of NPs
Eoil	Dielectric constant of oil
λ	Thermal conductivity [W.m <sup>-1</sup> .K <sup>-1</sup> ]
ρ	Density [g.cm <sup>-3</sup> ]
σ	Electrical conductivity [S.m <sup>-1</sup> ]
$\sigma_1$	Chapter 4, electrical conductivity of base fluid $[S.m^{-1}]$ Chapter 7, Charge density of NPs without surfactant polarization $[C \cdot m^{-3}]$
$\sigma_2$	Chapter 4, electrical conductivity of base fluid $[S.m^{-1}]$ Chapter 7, Charge density of NPs with surfactant polarization $[C \cdot m^{-3}]$

φ	Volume concentration of NPs
ω	Phonon frequency [rad/s]
τ	Relaxation time [s]
$ au_i$	Relaxation time of branch $i$ [s]

# Subscripts



### CHAPTER 1

### **INTRODUCTION**

### 1.1 Background

Insulating fluids based on mineral oil are used in oil-immersed power transformers. Mineral oil is a low-cost fluid that mostly accomplishes dual purposes, as a successful dielectric material and as a cooling medium (Committee of the IEEE Power & Society, 2018). However, the sustainable development of transformer oil is heatedly under debate in the world, because of the limitation of the resources of mineral oil, which mainly are based on petroleum products. Owing to sustainability, biodegradability and pollution-free nature of natural ester insulation oils, many researchers and industries investigated such oils for transformer applications (Rubalya Valantina et al., 2018; Salama et al., 2020; Turky & El-Adly, 2017).

In spite of vegetable oils could provide better properties as a transformer insulating oil; their commercial application faces some challenges. These challenges include the existence of unsaturated fatty acids and the extraction from edible products with a negative impact on food supply. To overcome this challenge, some additives were used with vegetable oils as antioxidants to enhance the critical properties of oil, especially under thermal aging conditions (Madavan & Balaraman, 2017; Raymon et al., 2013). Furthermore, the extraction of oil from natural esters those are not among edible oils.

The effective performance of natural ester fluids in an insulation system are governed by certain critical fluid properties that may influence the overall properties of the apparatus. It should have ample dielectric strength to cope with typical electrical stress in operation. It can be thermal conductive under a certain combination. Recently, the introduction of nanoparticles (NPs) was proposed to modify insulating fluids and prepare what is called nanofluids in order to improve thermal and dielectric properties (Geng et al., 2020; Huang et al., 2019; Minea, 2019; Qiu et al., 2020). The nanofillers are generally classified into metals, metal oxides and carbon-based substances. There are three types of NPs based on how many dimensions are in the nanometer scale. If the three dimensions are within the nanometer scale, the particle is considered zerodimensional. When two dimensions, the particle is considered one-dimensional. When single dimension, the particle is considered two-dimensional, NPs that are not confined to the nanoscale in any dimension having all three dimesions above 100 nm considered three-dimensional (Scher et al., 2016). In the current implementation two-dimensional graphene, GO, hexagonal boron nitride (h-BN) and zero-dimensional Al<sub>2</sub>O<sub>3</sub> were considered. The base fluid considered in the current research is non-edible cottonseed oil (CSO).

### **1.2 Problem statements**

Vegetable oils have emerged as prominent and effective alternative insulating fluids in transformer applications. Turning vegetable oil feedstock into a competitive commercial transformer grade insulating oil with better inherent properties imposes numerous practical consequences and challenges. Among them vital gridlock in most of the vegetable oils is that they are edible causing their application on a large scale to be limited (Ravulapalli et al., 2019). Accordingly, a non-edible vegetable oil insulating fluid is developed in this research. The nanofillers are generally classified into metals, metal oxides and carbon-based substances. Using metal oxides as nanofillers could improve the dielectric properties of the base fluids (Aljure et al., 2018; Fal et al., 2018). The h-BN or graphene NPs have a multifaceted feature among various 2D materials. However, h-BN overtakes other nanofillers and is an enticing material with high thermal conductivity and electrical insulation properties (Madavan et al., 2018; Yao et al., 2018). With respect to carbon-based nanofillers, they could improve thermal properties of the base fluids. Carbon nanotubes (CNTs) are coming at the forefront of these nanofillers, even at low weight percentage (wt%) (Fontes et al., 2015). These nanofillers have singledimensional morphology with Van der Waals  $\pi - \pi$  stacking interfaces, accelerating agglomeration between CNT fibrils. Aggregation and agglomeration are equivalent terms used by nanotechnology researchers to describe NPs that become clustered with each other in nanofluids. To avoid the aggregation of CNTs, they must be functionalized with strong acids, which induce accidental defects in uninterrupted sp2-bonded structures of carbon and significantly reduce thermal conductivity (Williams et al., 2010; Xuan et al., 2013). In chemistry, hybridization is defined as the process of combining two atomic orbitals to form a new kind of hybridised orbitals in a molecule. When one s and two p orbitals in the same shell of an atom combine to generate three analogous orbitals, this is referred to as sp2 hybridization (Peschel, 2011).

A stable nanofluid, in which NPs are homogeneously dispersed for a long-term, has steric repulsion that dominates Van der Waals attraction (Worthen et al., 2016). Few researchers investigated stability of DNFs (Dilectric nanofluids) for use in transformers (Avdeev et al., 2004; Dombek et al., 2014; Liu et al., 2011; Pislaru-Danescu et al., 2013; Shukla & Aiyer, 2015). Most works concluded that a low filler levels leads better stability than higher filler levels (Muhammad Rafiq et al., 2021), due to decreased probability of collision between NPs to form aggregates. It was also evident from these studies that visual inspection evaluation method was adopted to determine the dispersion stability. Visual inspection method is a qualitative method and is less effective; therefore quantitative Ultraviolet–visible (UV-Vis) spectroscopy method was adopted for the investigation of CSO based graphene nanofluids. The surfactant shields NPs, which prevents the formation of bonds with adjacent NPs when steric repulsion force dominates Van der Waals attraction. Some studies reported that addition of significant quantities of surfactant can shorten the dispersal time and weaken the dielectric properties of DNFs (Gupta et al., 2018; Rajnak et al., 2018).

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 NPs. This interfacial zone is considered to have a major impact on the dielectric properties of nanofluids (Mansour & Atiya, 2016), like its significance in solid nanodielectrics as reported by many researchers (Kochetov et al., 2012; Smith et al., 2008; Tanaka et al., 2005). 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. 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 (Hussain et al., 2020), 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 (Li & Li, 2019) based on S-parameter retrieval and considering the scattering of NPs. 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 (Miao et al., 2013), and could exhibit closer results to the experimental ones. However, all these models (Hussain et al., 2020; Li & Li, 2019; Miao et al., 2013) did not consider the surfactant polarization. NPs are generally anti-lipophilic and have a tendency to aggregate when they embedded in nanofluids. Thus, using of surfactants became a major process in achieving long-term dispersion stability of nanofluids through surface modification of NPs (Li et al., 2016; Muhammad Rafiq et al., 2021). So, it is crucial to consider the surfactant polarization in the relative permittivity calculation of nanofluids.

The following problem statements are identified from the above discussion:

- 1. Vegetable oils have emerged as prominent and effective alternative insulating fluids in transformer applications. However, most of vegetable oils are edible causing their application on a large scale to be limited (Ravulapalli et al., 2019).
- 2. Few researchers investigated stability of DNFs for use in transformers (Avdeev et al., 2004; Dombek et al., 2014; Liu et al., 2011; Pislaru-Danescu et al., 2013; Shukla & Aiyer, 2015) and adopted less effective qualitative visual inspection method (Primo et al., 2019).
- Relative permittivity models proposed earlier (Hussain et al., 2020; Li & Li, 2019; Miao et al., 2013) did not consider the surfactant polarization. Also, few researchers have examined the interfacial zone, focusing only on its influence on dispersion behaviour and stability (Mansour, Elsaeed & Izzularab, 2016).

## **1.3** Research aim and objectives

The aim of this study is to develop natural ester insulating nanofluids with enhanced dielectric and thermal properties. Several objectives have been set to achieve this aim.

1. To develop non-edible CSO as an insulating fluid with improved dielectric and thermal properties with the infusion of h-BN and GO NPs.

- 2. To enhance dispersion stability (through effective quantitative UV-Vis investigation method), dielectric properties and thermal properties of the graphene based CSO nanofluids by varying sonication time.
- 3. To design a relative permittivity model of composite fluids created by interfacial zone considering surfactant polarization that could exhibit closer results to the experimental results.

## **1.4** Scope of the research

The research activities associated with achieving each objective will be further elaborated in Chapters 4 to 7. The listed research activities for each research objective are clarified as follows:

### Scope of objective 1:

- To develop a non-edible CSO as an insulating fluid, CSO was selected as base insulating fluid. Tertiary butylhydroquinone (TBHQ) has been used as antioxidant, while h-BN and GO NPs used as nanofillers at varying weight percentage. h-BN was not modified with any surfactant while GO was modified with Sodium dodecyl sulfate (SDS) surfactant.
- NPs were comprehensively investigated for structural characterization. Twostep method was implemented for preparing nanofluids for the current implementation.
- Breakdown strengths under AC and lightning impulse voltages were evaluated. Also, other dielectric properties such as dielectric constant, dissipation factor, and resistivity were measured. In addition, thermal properties including thermal conductivity and thermogram analysis were obtained to investigate improved dielectric and thermal properties.
- The governing mechanism for enhanced dielectric properties were clarified by EDL (Electrical double layer), polarizability, and trapping charge carriers, while, thermal performance enhancement was clarified by phonon transport through nanosheets, Brownian motion, and EDL.

### Scope of objective 2:

- To investigate the effect of sonication time on dispersion stability of graphene based CSO nanofluids through quantitative UV-Vis spectroscopy method. CSO based DNFs were developed using a combination of graphene and sodium dodecylbenzene sulfonate (SDBS).
- First, experimental methods introduced are preparation of DNFs and investigation of short-term stability by visual inspection after two weeks.

- Based on short-term stability results, unlike previously published works two optimum best ultrasonication periods were used for long-term stability evaluation through UV-Vis spectroscopy, and dielectric and thermal properties investigation.
- Comparing stability, dielectric and heat transfer properties at these optimum sonication times. Finally, the governing mechanisms behind the obtained results were discussed.

### Scope of objective 3:

- Relative permittivity model of composite fluids considering surfactant polarization that could exhibit closer results to the investigated experimental results was designed referring to the Clausius-Mossotti equation with proper assumptions.
- CSO based nanofluids were developed using a combination of nanofiller  $Al_2O_3$  and surfactant SDBS through ultrasonication.
- Experimental relative permittivity results of DNFs at different concentrations and varying temperature were investigated.
- Relative permittivity results calculated from the model were compared with experimental relative permittivity results to check robustness of proposed model.
- A relative permittivity model of composite fluids considering surfactant polarization was proposed. Further, effects of interfacial zone on improved relative permittivity, AC BDV (Breakdown Voltage), and dissipation factor were discussed with the help of obtained results.

### 1.5 Contribution of the research

Details of the research's contributions are as follows:

- 1. Non-edible CSO was proposed as natural ester insulating fluid through inclusion of antioxidant and filling with h-BN nanofillers. All the obtained results and clarified mechanisms in the present study validate the superiority of CSO based h-BN nanofluids as a potential candidate either for power equipment or for management thermal energy systems.
- 2. Natural esters proved beneficial performance considering some practical issues. For example, papers impregnated in natural esters had higher tensile strengths than that impregnated in mineral oil under various aging conditions. Also, natural esters could prove lower annual percent loss of life compared to mineral oil. In addition to these properties in the current research, the 2D carbon material GO nanosheets' nature contributes to elevated electrical insulation, making them attractive nanofillers for improving the transformer oil thermal properties.

- 3. The effect of sonication time on dispersion stability, dielectric properties, and heat transfer of graphene based CSO nanofluids was investigated. The presented results showed the relation between dispersion stability and thermodielectric properties. This contributes to the key issue of the long-term stability of the natural ester graphene nanofluids when working within a transformer.
- 4. The surfactant can have a crucial impact on the interfacial zone of NPs to improve dielectric properties of nanofluids under the application of an external electrical field. Accordingly, surfactant polarization model of composite fluids is proposed in this research to calculate relative permittivity of nanofluids. This helps researchers to calculate gross relative permittivity of nanofluids before actual development.

### **1.6** Organization of the thesis

The study is structured in eight chapters to achieve the above mentioned objectives in the UPM alternative thesis format of thesis by publication (style 3). The full-length articles covering objectives are chapters 4, 5, 6 and 7.

Chapter 1 provides the introduction of current research. The background, problem statement, objectives, scope, and the research contribution were discussed briefly.

Chapter 2 starts with the introduction followed by concerns about the commercially available liquid dielectrics and motivations behind using natural ester oil as an alternative. In addition, the advantages of natural ester insulating fluids are discussed with highlighting their chemical composition and percentage of fatty acids. Further, challenges related to natural ester insulating fluids are discussed for real time applications; methods to improve the effectiveness of natural ester insulating oils are discussed in detail in the effort to meet these associated challenges. Among these methods is the concept of nanofluids. Therefore, an extensive review has been presented to improve the dielectric and thermal properties of natural ester insulating oils through proposing novel natural ester based nanofluids. Furthermore, effect of sonication time on stability and thermo-electric properties of graphene nanofluids were reviewed. Finally, the effect of interfacial zone due to NP-surfactant interaction on dielectric properties of nanofluids was presented extensively. To calculate relative permittivity of nanofluids existing relative permittivity models were discussed. As a result of this extensive review, gaps are identified to address them in further chapters.

The chapter 3 explains brief methodology adopted to achieve all three objectives. The experimental investigation began by characterization of research-grade NPs (i.e., h-BN, GO, graphene, and  $Al_2O_3$ ) and then by preparing the base CSO. The first step in achieving all three objectives was the preparation of h-BN, GO, graphene and  $Al_2O_3$  nanofluids at different filler levels. This is followed by experimental evaluation for stability analysis of prepared nanofluids through UV-Vis/Visual inspection, electrical

and/or thermal measurements. Then, the test results were validated. In order to achieve objective 3 together with the above steps, proposed mathematical model results were compared with experimental results in order to check its robustness.

The chapter 4 deals with the first objective supported by published research article entitled "Investigation of the dielectric and thermal properties of non-edible CSO by infusing h-bn NPs". This study aimed to propose non-edible CSO as natural ester insulating fluids through filling with h-BN nanofillers for four dosage levels (0.01, 0.02, 0.05, and 0.1 wt%). Breakdown strengths under AC and lightning impulse voltages, dielectric constant, dissipation factor, resistivity, and thermal properties were presented.

The chapter 5 again addresses the first objective supported by published article entitled "Development of graphene oxide-based nonedible cottonseed nanofluids for power transformers". This study aimed to propose dielectric nanofluids with graphene oxide nanofillers as two-dimensional (2D) hexagonal carbon lattice nanofillers at four weight percentages (0.01, 0.02, 0.03 and 0.05 wt%). The dielectric and thermal properties of nanofluids were evaluated in comparison to base CSO.

The chapter 6 addresses the second objective supported by published article entitled "Investigation of the effect of sonication time on dispersion stability, dielectric properties, and heat transfer of graphene based green nanofluids". This study aims to improve dispersion stability (through quantitative UV-Vis investigation method), dielectric properties and thermal properties of the graphene based CSO nanofluids by varying sonication time. Based on short-term stability results, two ultrasonication periods were used for preparing nanofluids (0.0015, 0.003, 0.006, and 0.01 wt%) for long-term stability evaluation through UV-Vis spectroscopy. For the two most stable nanofluids, stability, dielectric and thermal properties were investigated and compared.

The chapter 7 addresses the third objective supported by submitted article entitled "The effect of interfacial zone due to NP-surfactant interaction on dielectric properties of vegetable oil based nanofluids". This research focuses on to propose a relative permittivity model of composite fluids considering surfactant polarization that could exhibit closer results to the experimental results. Experimental relative permittivity results of nanofluids at different concentrations (0.01, 0.02, 0.03, and 0.05 wt%) and varying temperature (Viz., 45 °C, 60 °C, 75 °C and 90 °C) were investigated and compared with proposed relative permittivity model. Finally, the role of interfacial zone on dielectric properties of CSO based Al<sub>2</sub>O<sub>3</sub> nanofluids was investigated.

Finally, chapter 8 summaries the conclusions of the research study as well as the recommendations for future work.

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