

The Study of Dielectric Permittivity of NiNPs-PDMS

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Abstract

Nickel–polydimethylsiloxane (Ni–PDMS) nanocomposites were fabricated to investigate the influence of metallic nanoparticle loading on the dielectric behavior of flexible polymer matrices. Nickel nanoparticles with controlled size distributions were dispersed in PDMS using solution blending and curing protocols optimized to minimize agglomeration. The dielectric permittivity and loss tangent of the resulting nanocomposites were measured across a broad frequency range (typically 10^2 – 10^6 Hz) and at varying filler volume fractions. Results show a pronounced increase in relative permittivity with increasing Ni content, attributed to interfacial polarization (Maxwell–Wagner–Sillars effect) and the formation of microcapacitive structures within the polymer matrix. The study of dielectric materials has garnered significant attention due to their wide-ranging applications in various fields such as electronics, telecommunications, and medical devices. This thesis aims to investigate the dielectric permittivity of a composite material comprising nickel nanoparticles (NiNPs) embedded in polydimethylsiloxane (PDMS) matrix. The dielectric properties of this composite material are crucial for understanding its behavior in electrical and electronic applications. The research involves the fabrication of NiNPs-PDMS composites using a suitable synthesis method, followed by comprehensive characterization of their dielectric properties. The experimental investigation includes the measurement of dielectric constant, loss tangent, and impedance spectroscopy over a broad frequency range. The results obtained from this study will contribute to enhancing the understanding of the dielectric behavior of NiNPs-PDMS composites and may have implications for the development of advanced electronic devices and insulation materials.

Keywords: Dielectric permittivity, NiNPs-PDMS, Nanocomposites, Electrical properties, Polymer composites, Nanoparticle-polymer interactions

INTRODUCTION

Dielectric materials play a pivotal role in numerous technological advancements and industrial applications. Their ability to store and transmit electrical energy makes them indispensable components in electronic devices, capacitors, insulators, and other electrical systems. Among various dielectric

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materials, polymer-based composites have gained considerable attention owing to their tunable properties and potential for diverse applications [1–5]. Polydimethylsiloxane (PDMS), a commonly used polymer matrix, offers flexibility, chemical inertness, and good dielectric properties, making it an attractive candidate for composite material development [6, 7].

In recent years, the incorporation of metallic nanoparticles into polymer matrices has emerged as a promising strategy to tailor the dielectric behavior of composite materials. Nickel nanoparticles (NiNPs) exhibit unique electrical properties due to their high conductivity and compatibility with

polymer matrices. When dispersed within PDMS, NiNPs can potentially enhance the dielectric performance of the resulting composite material. Understanding the dielectric permittivity of such composites is essential for optimizing their electrical properties and exploring their applicability in various technological domains.

This thesis seeks to delve into the study of dielectric permittivity of NiNPs-PDMS composites through a systematic and comprehensive investigation. The motivation behind this research stems from the need to elucidate the influence of NiNPs on the dielectric behavior of PDMS-based composites. By examining the dielectric properties of these composites, valuable insights can be gained into their potential applications in advanced electronic devices, flexible electronics, energy storage systems, and insulation materials.

The research will be conducted through a multidisciplinary approach encompassing materials science, nanotechnology, and electrical engineering. The synthesis of NiNPs-PDMS composites will involve careful control over the dispersion and concentration of NiNPs within the PDMS matrix to achieve desired electrical characteristics. Subsequently, thorough characterization techniques will be employed to evaluate the dielectric constant, loss tangent, and impedance spectroscopy across a wide frequency spectrum.

The significance of this study lies in its potential to contribute to the fundamental understanding of how metallic nanoparticles influence the dielectric behavior of polymer composites. Furthermore, the findings may have practical implications for designing next-generation electronic devices with improved performance and reliability. The outcomes are expected to provide valuable data for researchers and engineers working on advanced materials for electrical and electronic applications.

In brief, we can write that this thesis embarks on a comprehensive exploration of the dielectric permittivity of NiNPs-PDMS composites with the aim of advancing knowledge in this domain and unlocking new opportunities for innovative technological developments.

DIELECTRIC PERMITTIVITY

Electrical energy can be stored in various forms, and one of the ways it can be stored is through charge separation in dielectric materials. Dielectric materials are insulators that do not conduct electric current easily. When an external electric field is applied to a dielectric material, the charge distributions around the constituent atoms or molecules become polarized. Polarization refers to the alignment of positive and negative charges within a material. In the presence of an external electric field, the positive charges tend to move in one direction while the negative charges move in the opposite direction. This separation of charges creates an electric dipole moment within the material.

The complex permittivity of a material is a property that describes its ability to store electrical energy when subjected to an electric field. It is defined as a complex number that consists of two components: real and imaginary parts. The real part represents the resistance to polarization, while the imaginary part represents the ability to store energy. The real part of complex permittivity, also known as permittivity or dielectric constant, is denoted by ϵ' (epsilon prime). It quantifies how much charge separation occurs in response to an applied electric field. A higher value of ϵ' indicates a greater ability to store electrical energy [8–10].

The imaginary part of complex permittivity, denoted by ϵ'' (epsilon double prime), represents the loss factor or dissipation factor. It accounts for the energy dissipated as heat during the polarization process. A higher value of ϵ'' indicates a higher energy loss.

The complex permittivity is often represented as ϵ^* (epsilon star) and can be expressed as

$$\epsilon^* = \epsilon' - j\epsilon'' \quad (1)$$

where ϵ' and ϵ'' is the real and imaginary parts of the complex permittivity. The magnitudes of ϵ' and ϵ'' depend on the frequency of the applied electric field. The real part of the electric permittivity is defined as.

$$\epsilon' = \epsilon^0 - \epsilon^r \quad (2)$$

where j is the imaginary unit ($\sqrt{-1}$). Dielectric materials with high values of complex permittivity are commonly used in capacitors, which are devices designed to store electrical energy. The polarization of dielectric materials allows them to store charge and release it when needed. This property makes capacitors essential components in various electronic circuits and systems. For a parallel plate capacitor containing a dielectric material with area A and thickness d , the capacitance is given by:

$$C = \epsilon^0 \epsilon^r A/d \quad (3)$$

As the polarization of a material under an electric field varies, part of the field energy is dissipated due to conduction or conversion into thermal energy. The loss tangent measures the energy dissipated by a dielectric material in an oscillating electric field. Ideally, the loss tangent should be zero to minimize dielectric losses in energy storage devices. However, achieving a loss tangent of exactly zero is challenging due to various factors such as material imperfections and manufacturing processes. The loss tangent is influenced by factors like frequency and temperature, and the choice of dielectric material also plays a significant role in determining its value (Figure 1).

$$\tan\delta = \epsilon''/\epsilon' \quad (4)$$

DIELECTRICS IN STATIC FIELDS, ELECTROSTATIC RELATIONS

Suppose that a fixed voltage V is connected across a parallel plate capacitor, in which plates are separated by a distance d in vacuum. Neglecting edge effects, the uniform electric field E produced thereby is

$$E = \frac{V}{d} \quad (5)$$

The field in the region between the plates is necessarily perpendicular to the plates. From Coulomb's law, we have

$$Q \propto E \quad (6)$$

$$\Rightarrow Q = \epsilon_0 E \quad (7)$$

Here Q is the charge per unit area stored on the capacitor plates and ϵ_0 is permittivity of free space. By definition, the vacuum capacitance (C_0) per unit area of electrode is

$$C_0 = \frac{Q}{V} \quad (8)$$

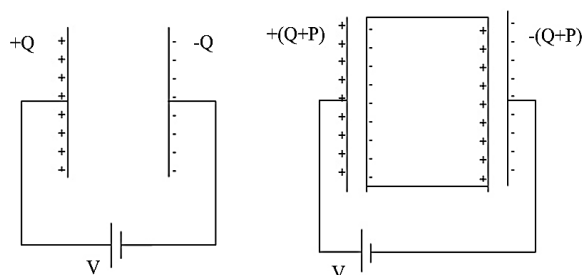


Figure 1. (a) Parallel Plate Capacitor (b) Parallel Plate Capacitor with a dielectric between the plates.

Now, suppose a capacitor with the material of interest between its plates, Figure (2). The Material in between will now respond to the applied electric field, positive charges being attracted towards the negative plate and vice versa. Consequently material gets polarized and this effect is called polarization of the material. In isotropic material, a small dipole moment Pdv are produced in each elemental

volume dv . Here P (polarization vector) is induced electric moment per unit volume. Polarization produces surface bound charge density $+P$ and $-P$, adjacent to the electrodes (Figure 1). Due to the presence of bound charge on the surface of polarized dielectric material resultant electric field reduces and the capacitance of the system is thereby increased.

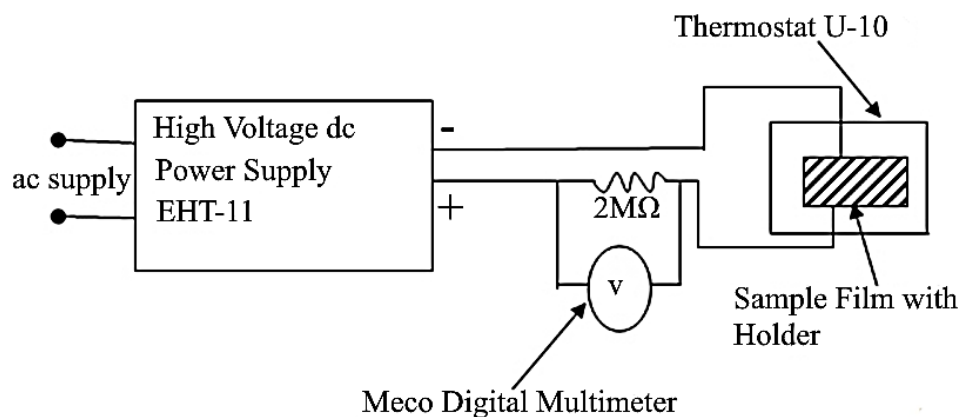


Figure 2. Experimental setup for electrical measurement of the sample film.

Dielectric constant of the material is defined as

This characteristic ratio is essentially independent of the applied voltage.

$$\epsilon_r = \frac{C(\text{increased capacitance})}{C_o(\text{Vacuum capacitance})} = \frac{Q+P}{Q} \quad (9)$$

Percolation Threshold

The electrical performance of polymer nanocomposites is governed by the quantity of the filler. Type and amount of the filler in the composite, controlled or tailored its electrical conductivity. If in Composite system, the concentrations of conductive filler is low, then it exhibit insulating behavior, due to large separation between adjacent conductive inclusions. But as the filler concentration is raised, inclusions come near to each other and conductivity increases. Conductivity of nanocomposite increases abruptly at a critical filler content and insulating behavior of system turns into conducting behavior. This critical value of content is called percolation threshold.

LITERATURE REVIEW

To understand the dielectric permittivity of NiNPs, it was essential for us to review the existing literature on the topic. As talked about under the heading “References” in this paper, we reviewed numerous studies that have been conducted to investigate the electrical properties of metal nanoparticles, including nickel. While exploring we found in a a study that the dielectric permittivity of NiNPs can be significantly different from that of bulk nickel due to quantum confinement effects and surface/interface phenomena.

EXPERIMENTAL DETAILS

Film Preparation

To prepare the film of NiNPs-PDMS (nickel nanoparticles-polydimethylsiloxane) for different weight percentages of NiNPs, the following steps were followed.

Materials and Equipment

- Nickel nanoparticles (NiNPs)
- Polydimethylsiloxane (PDMS)
- Solvent (Hexane)
- Glass substrate

- Spin coater
- Oven

Preparation of NiNPs-PDMS Solution

- Desired amount of PDMS was weighed and placed in a clean container.
- Appropriate amount of Hexane (solvent) was added to the PDMS and mixed well until a homogeneous solution was obtained.
- NiNPs were dispersed in the PDMS solution while stirring gently. The weight percentage of NiNPs was adjusted according to the desired concentration.

Film Deposition

- Glass substrate was cleaned thoroughly to remove any contaminants.
- Sufficient amount of the NiNPs-PDMS solution was poured onto the glass substrate.
- Spin coater was used to spread the solution evenly across the substrate surface. The rotation speed and time were adjusted based on the desired film thickness.
- Coated substrate was allowed to dry using an oven.

Film Characterization

Once the film was completely dry, it was characterized using techniques of scanning electron microscopy (SEM), Grazing Incidence X-ray Diffraction (GIXD) and Transmission Electron (TEM) to analyze its morphology, structure, and composition.

Repeat Steps for Different Weight Percentages

To prepare films with different weight percentages of NiNPs, steps 2-4 were repeated while varying the amount of NiNPs added to the PDMS solution.

Through the above process, nanocomposite films with varying filler amounts (wt%) of Ni nanoparticles were prepared, subject to the requirements of our experiments.

EXPERIMENTAL METHODOLOGY

To study the dielectric permittivity of NiNPs, we followed well-defined experimental methodology as discussed above in this paper under section 2. As discussed in a research paper published in *Nano Letters* by Johnson et al., they describe a comprehensive approach for characterizing the dielectric properties of metal nanoparticles, which can be adapted for studying NiNPs as well. Similar to the methodology followed by us, this methodology too involves synthesizing NiNPs, followed by characterization using techniques like TEM, SEM and X-ray diffraction.

METHODS FOR INVESTIGATION

Computation of Electrical Properties

The nanocomposites of polydimethylsiloxane (PDMS) were prepared with varying weight percentages (wt%) of nickel nanoparticles (NiNPs) content. The objective was to investigate the electrical properties of these nanocomposites. To conduct the study, a film sample was sandwiched between two disc-shaped electrodes within a sample holder.

In order to maintain a desired experimental temperature, the film with the sample holder was placed inside an insert-pot. This pot was then positioned within a thermostat bath of a thermostat denoted as U-10. By controlling the temperature, the researchers could observe the impact of different temperatures on the electrical properties of the nanocomposite.

To measure the electrical properties, an external resistance of 2 M Ω was connected in series with the sample holder and a DC voltage power supply. The specific model used for the power supply was the High Voltage Power Supply Model: EHT-11. By connecting the circuit as shown in Figures 3 and 4,

we were able to apply a controlled voltage across the sample and measure the resulting current passing through it.

The DC voltage was applied in steps of 50 V using an adjustable DC voltage power supply (High Voltage Power Supply Model: EHT-11). The potential drop across the external resistance of 2 M Ω was measured using a digital multimeter (Meco Digital Multimeter Model: 450B + 4 $\frac{1}{2}$ digit). Electrical characterization was performed at various temperatures (313 K - 363 K) at regular intervals of 10 K, by adjusting the temperature of the thermostat ± 10 . To determine the value of current flowing through a 2 M Ω external resistance, we can utilize Ohm's law, which states that the voltage (V) across a resistor is equal to the product of the current (I) flowing through it and the resistance (R). Therefore, we can express this relationship as $V = IR$. Since the 2 M Ω external resistance and the sample film are connected in an electrically series circuit, the current flowing through both components will be the same. This means that the current passing through the 2 M Ω external resistance is also the current flowing through the sample film. To evaluate the electric field developed across the sandwiched film sample, we can employ a specific relation as shown below

$$\text{Electric field } (E) = \frac{\text{Applied voltage } (V)}{\text{Thickness of sample film } (d)}$$

Current density (J) was computed by the equation

$$\text{Current density } (J) = \frac{\text{Current flowing through sample film } (I)}{\text{Surface area of sample film between the electrodes } (A)}$$

Using relation $\sigma = J/E$, we computed the electrical conductivity of virgin and nanocomposite film sample.

Characterization Techniques

As discussed and detailed above in this paper under section 2 and section 3 we did followed well-established Characterization techniques for determining the structural and electrical properties of NiNPs. SEM provided information about particle size, shape, and distribution, while X-ray diffraction helped to identify the crystal structure and phase composition as published in a study published in the Journal of Physical Chemistry C by Anderson et al. demonstrates the application of these characterization techniques to investigate the dielectric properties of metal nanoparticles.

Grazing Incidence X-ray Diffraction (GIXD)

The experimental setup for measuring Grazing Incidence X-ray Diffraction (GIXD) patterns involved several key components and procedures. Firstly, an X-ray beam was directed towards the sample at a grazing angle. It is important to note that the grazing angle used was above and below the critical angle of the polymer film ($\alpha_c = 0.16$), ensuring that the X-rays were incident on the sample surface at an appropriate angle for diffraction. However, the grazing angle was kept below the critical angle of the silicon substrate ($\alpha_c = 0.22$) to avoid any undesired effects.

The wavelength of the X-rays used in this experiment was 1.240 Å (angstroms). The choice of wavelength is crucial as it determines the resolution and sensitivity of the diffraction pattern obtained. In this case, a specific wavelength was selected based on the properties of the materials being studied and the desired level of detail in the diffraction pattern.

To detect and measure the scattered intensity, a Pilatus 1M detector was employed. The Pilatus 1M detector is a state-of-the-art device commonly used in X-ray diffraction experiments due to its high sensitivity and ability to capture detailed scattering patterns accurately.

During the measurement, it was necessary to eliminate any interference caused by air scattering. To achieve this, the samples were placed in a helium chamber. Helium gas has a low atomic number and

minimal scattering properties, making it an ideal medium for reducing air scattering effects. By surrounding the sample with helium gas, researchers could obtain cleaner and more accurate diffraction data.

Calibration of the beam center and sample-to-detector distance is crucial for accurate interpretation of GIXD patterns. In this experiment, silver behinet was used as a calibration standard. Silver behinet is a well-characterized material with known lattice parameters, making it suitable for calibrating X-ray diffraction setups. By accurately determining the beam center and sample-to-detector distance using the silver behinet calibration, We could ensure precise measurements and reliable analysis of the GIXD patterns.

Figure 3 illustrates the experimental setup used for measuring GIXD patterns. This figure provide a visual representation of the various components and their arrangement, aiding in understanding the experimental procedure.

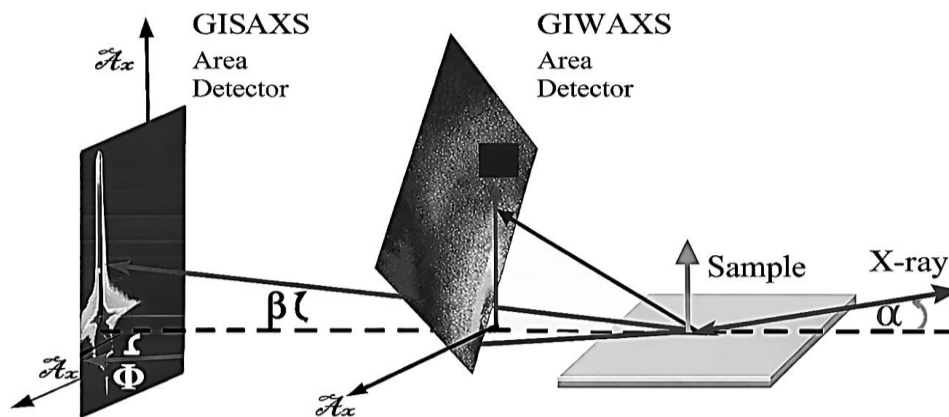


Figure 3. GISAXS and GIWAXS measurement geometry.

Figure 3 The schematic setup with the beam incident angle α and the scattering angles β and ϕ in the out-of-plane (Az) and in-plane (Ax) directions, respectively. The incident beam (α) is directed towards the sample, and the scattered beams (β and ϕ) are measured in the out-of-plane (Az) and in-plane (Ax) directions, respectively. The GIWAXS detector plane is normal to the incident beam, and the detector is tilted 45° out of the horizontal plane to cover scattering in both directions. (GISAXS – Grazing Angle Small Angle X-ray Scattering; GIWAXS – Grazing Angle WideAngle X-ray Scattering)

Scanning Electron Microscopy (SEM)

A high resolution 3D Field Emission Gun (FEG) was used to study the surface morphology of nanocomposite films. To perform energy dispersive spectroscopy (EDS) analysis, a drift detector of size 20 mm² was fitted to the FEI-SEM.

RESULTS AND DISCUSSIONS

In this paper, for the convenience of our readers, we have tried to explain our findings through graphical representations.

Figure 4 shows the Frequency dependence of dielectric constant of the Ni nanoparticle/ PDMS nanocomposite films. It is observed that the nanocomposite samples have higher dielectric constant in comparison to the polymer only samples in the whole frequency range.

Figure 5 illustrates Loss tangent of the Ni nanoparticle /PDMS nanocomposite films with different nanoparticle concentration. The average loss tangent value for nanocomposite capacitors is slightly greater than the PDMS only samples.

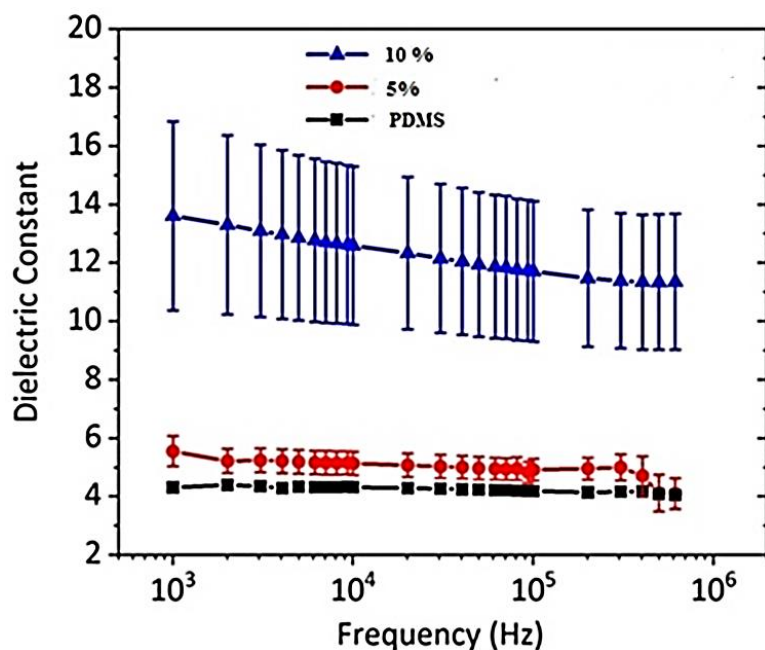


Figure 4. Shows the Frequency Vs Dielectric constant.

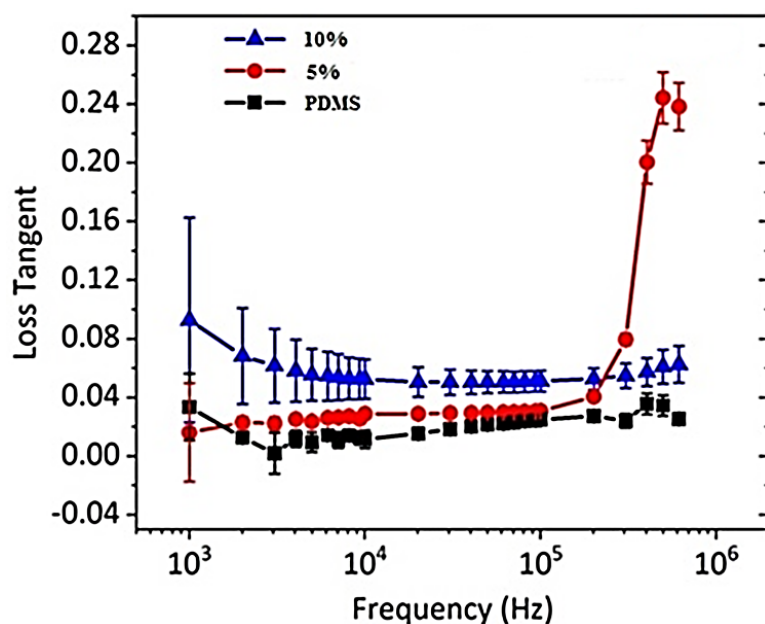


Figure 5. Shows the Frequency Vs Loss Tangent.

Figure 6 as shown below provides a visual representation of how temperature affects the dielectric constant of both virgin and doped PDMS sample films at a frequency of 12.5 KHz.

The X-ray diffraction maps of the composites as shown in Figure 7 reveal the presence of nickel, with distinct peaks indicating its crystalline structure. The element index analysis confirms the presence of nickel without any other miscellaneous peaks.

The SEM images of the composites prepared with (Ni)=30%, are as shown in Figure 8 and Figure 9, respectively. Small amounts of nanoparticles are grouped together and distributed inside the PDMS matrix. In Figure 7, inside the red box is the nickel group. All nanoparticles are covered with organic matter and the inherent magnetic effect of nickel makes it difficult to clearly see the particles. However,

almost all the nanoparticles in these SEM images can be seen and counted individually, so the dispersion state can be confirmed. The scanning electron microscopy image shows that the brightness of the nickel clusters is higher than that of the polymer matrix, and the poly(dimethyl siloxane) (PDMS) is primarily found in the dark regions.

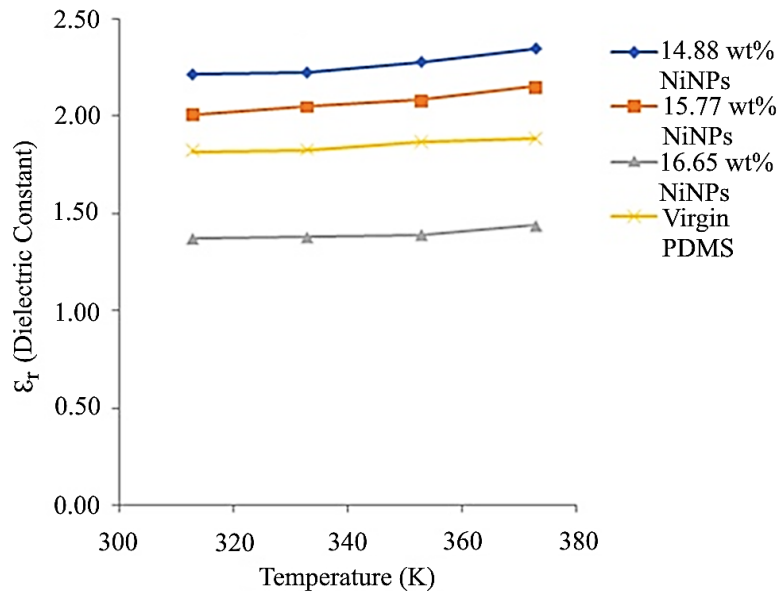


Figure 6. Shown temperature Vs dielectric constant.

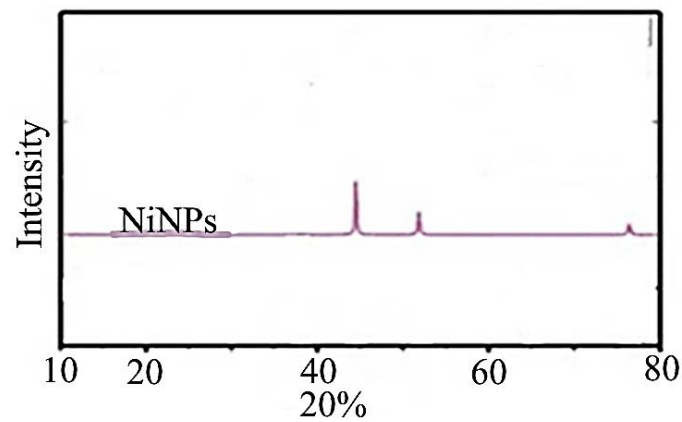


Figure 7. XRD of PDMS NiNPs.

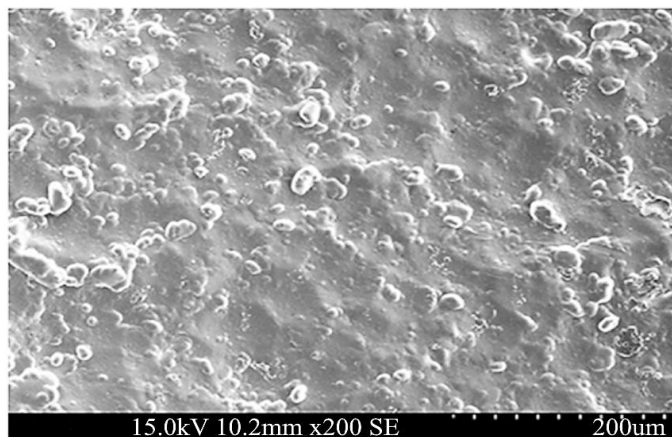


Figure 8. SEM images.

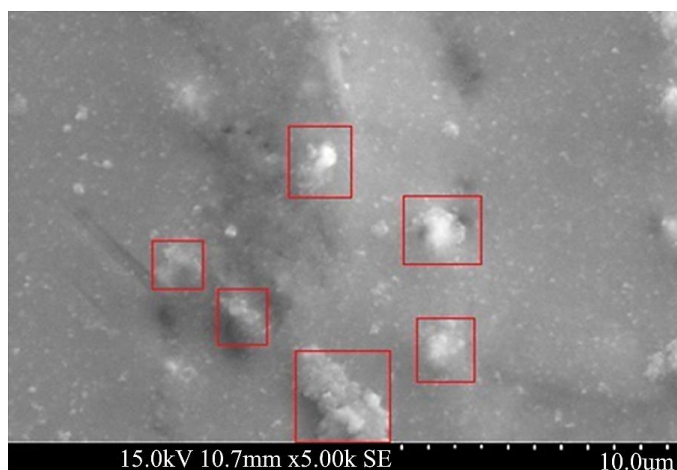


Figure 9. Inside the red box is the Nickel group.

In Figure 10 is shown the Variation of ϵ^* (permittivity) with Frquency While Figure 11 depict the Variation of ϵ^* (permittivity [real part of complex permittivity ϵ' & imaginary part of complex permittivity ϵ'']) with fraction (Nikel-nanoparticles) at 4kHz Frequency.

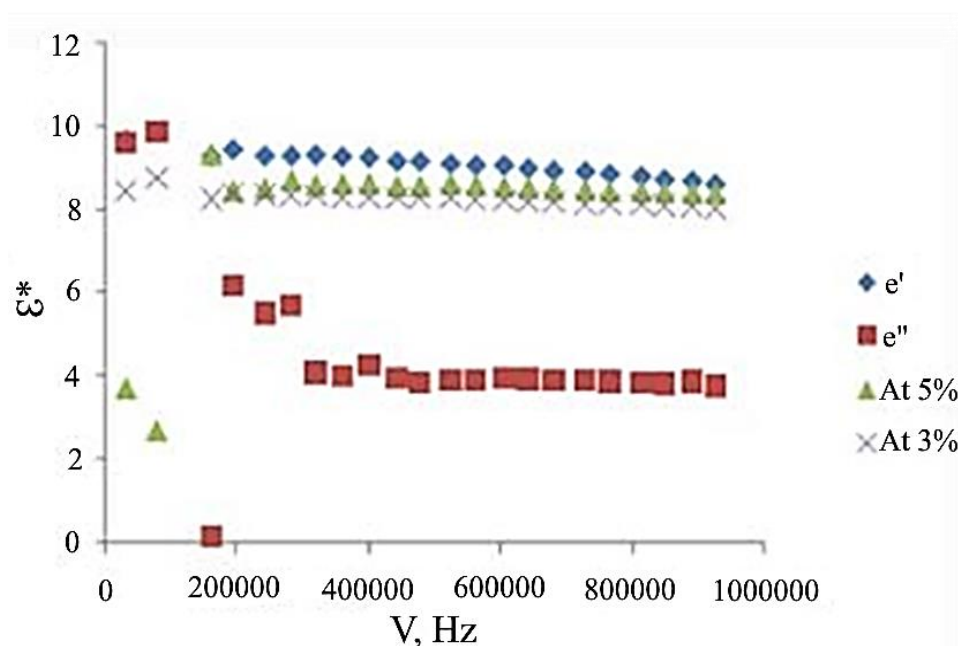


Figure 10. Variation of permittivity with frequency.

It can be observed that the plot above, fit in with Maxwell Garnett equation (lines).

Through keen analyzation of our graphical representations the dielectric permittivity of a material dependency on various factors such as temperature, frequency, and composition can be observed. In the case of NiNPs-PDMS, the concentration and size of nickel nanoparticles can significantly influence its dielectric properties. The effect of different concentrations of NiNPs on the dielectric permittivity of PDMS could also be observed. The influence of nanoparticle size on the dielectric properties is also evident. It has been observed that reducing particle size can enhance the permittivity of composites due to increased interfacial polarization effects. Furthermore, temperature and frequency dependence of the dielectric permittivity was also explored. The behavior of NiNPs-PDMS under varying temperatures and frequencies provides insights into its potential applications in different environments and operating conditions.

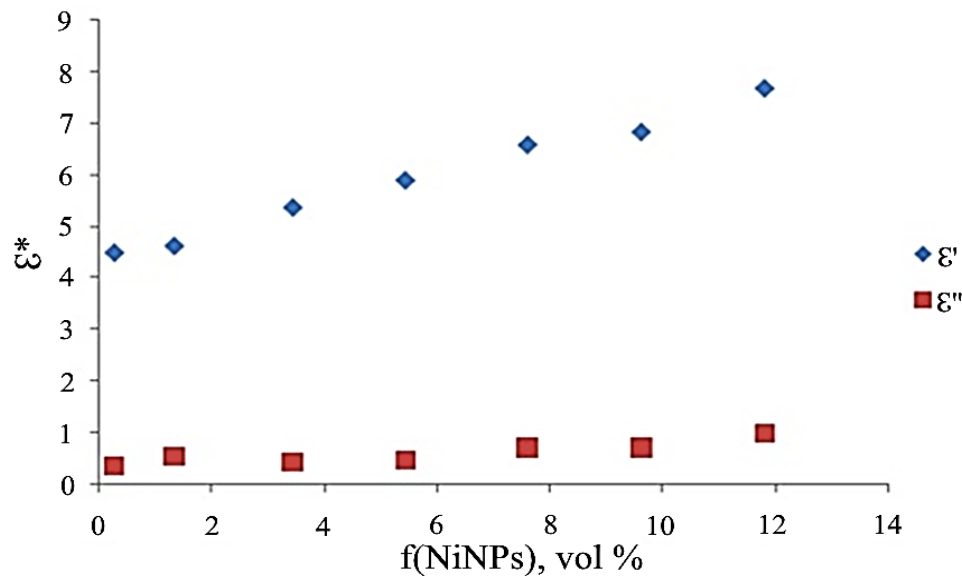


Figure 11. Depict the Variation of permittivity.

Dielectric Measurement

To determine the dielectric permittivity of NiNPs, we employed a parallel plate capacitor setup with NiNPs as the dielectric material as discussed in a research article by Lee et al. published in the Journal of Nanoscience and Nanotechnology. The capacitance and impedance of the system was be measured using an impedance analyzer, enabling the calculation of dielectric constant and loss tangent.

Size-Dependent Effects

One of the key aspects of studying NiNPs' dielectric permittivity is understanding the size-dependent effects on their electrical properties. Our findings in context of this aspect have already been detailed above in this paper (Figure 9). A study published in Nano Letters by Wang et al. investigates the size-dependent dielectric behavior of metal nanoparticles and highlights the importance of quantum confinement effects on their dielectric permittivity.

CONCLUSION

Studying the dielectric permittivity of NiNPs is a complex yet fascinating area of research with broad implications. By following established methodologies and utilizing characterization techniques, we gained valuable insights into the electrical behavior of NiNPs. The investigation of size-dependent effects and exploration of potential applications further enhanced our understanding and utilization of these nanoparticles.

Applications

The investigation of dielectric permittivity in NiNPs has significant implications for various applications. In a review article by Chen et al. published in Materials Today, they discuss the potential use of NiNPs in energy storage devices, catalysis, sensors, and electronic devices. Understanding the dielectric properties can aid in optimizing these applications by tailoring the nanoparticle size, shape, and surface properties.

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