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REGULAR ARTICLE

Effect of Temperature on Dielectric and Impedance Characteristics of Graphene Oxide Thin Films

Shib Shankar Biswas[1], Hari Shankar Biswas[1,*], Dilip Kumar Maiti[2,†]

[1] *Department of Chemistry, Surendranath College, 700009 Kolkata, India*
[2] *Department of Chemistry, University of Calcutta, University College of Science, Kolkata-09, India*

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Graphene oxide (GO) thin films have attracted significant attention due to their unique combination of polar oxygen functional groups and a partially restored sp^2 carbon network, which makes their electrical behavior highly dependent on temperature and frequency. Unlike pristine graphene, GO exhibits a heterogeneous structure in which localized charge carriers, structural defects, and interfacial effects strongly influence its dielectric and impedance characteristics. These features make GO thin films promising for applications in sensors, flexible electronics, energy storage devices, and resistive switching systems, where temperature-dependent electrical properties play a crucial role. This paper presents a practical framework for understanding temperature-tuned dielectric spectroscopy and impedance behavior of GO thin films deposited on insulating substrates. By analyzing complex permittivity, electric modulus, and equivalent circuit models, we relate the observed frequency dispersion of dielectric constant (ε') and dielectric loss (ε'') to microscopic mechanisms such as Maxwell-Wagner interfacial polarization, charge hopping, and thermally activated transport. With increasing temperature, relaxation processes become faster, shifting dielectric loss peaks to higher frequencies and reducing overall resistance. Nyquist plots exhibit shrinking semicircles at elevated temperatures, indicating enhanced charge mobility and partial reduction of GO. The DC conductivity follows Arrhenius-type behavior, confirming thermally activated conduction.

Keywords: Graphene oxide, Impedance, Conductivity, Permittivity, Mobility, Electric modulus, Dielectric loss.

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1. INTRODUCTION

Graphene oxide (GO) is a chemically modified derivative of graphite that is produced through the controlled oxidation of crystalline graphite using strong oxidizing agents such as potassium permanganate in acidic media [1]. During this process, a high density of oxygen-containing functional groups-including epoxide (C-O-C), hydroxyl (-OH), carbonyl (C=O), and carboxyl (-COOH) groups are introduced onto the basal planes and edges of the graphene layers. These functional groups break the continuous conjugated sp^2 carbon network of pristine graphite and graphene, leading to significant structural disorder, lattice distortion, and electronic localization. As a result, GO behaves as an electrical insulator at room temperature, in contrast to highly conductive graphene. However, the presence of these polar functional groups also makes GO an extremely polar, hydrophilic, and chemically reactive material with a complex heterogeneous microstructure. At the microscopic level, GO consists of regions of oxidized sp^3 carbon domains interspersed with small islands of un-oxidized sp^2 carbon clusters. This hybrid structure gives rise to spatial inhomogeneity in electrical and dielectric properties, where localized conducting pathways are embedded within an insulating matrix [2]. The degree of oxidation, defect density, and distribution of functional groups strongly influence the overall electrical response of GO. Moreover, the presence of interlayer water molecules and residual ions trapped between GO sheets further contributes to its complex electrochemical and dielectric behavior.

When GO is fabricated into thin films, typically by techniques such as drop casting, spin coating, or spray coating, the material forms a stacked layered structure composed of overlapping and randomly oriented GO flakes. These stacked layers create numerous internal interfaces between adjacent sheets, which act as barriers to charge transport. Under an applied alternating electric field, charge carriers accumulate at these internal boundaries, leading to strong Maxwell-Wagner-Sillars (MWS) interfacial polarization, particularly at low frequencies [3]. This phenomenon arises from the contrast in conductivity and permittivity between the more conductive sp² regions and the highly insulating oxidized domains. Additionally, at the electrode-GO interface, charge accumulation results in electrode polarization effects, which further enhance low-frequency dielectric response and impedance dispersion. Temperature plays a crucial role in modulating the electrical and dielectric properties of GO thin films. At low temperatures, dipolar motion of oxygen functional groups and trapped water molecules is limited, resulting in slow relaxation

* Correspondence e-mail: harishankarb7@gmail.com
† dkmchem@univ.ac.in

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processes and high electrical resistance [4]. As temperature increases, thermal energy enhances the mobility of dipoles and facilitates charge carrier movement through hopping conduction mechanisms. In this regime, electrons or holes can hop between localized sp^2 clusters, while protons and ions can migrate through hydrogen-bonded networks associated with interlayer water and functional groups. This thermally activated transport leads to a gradual decrease in resistivity and an increase in ac and dc conductivity with temperature.

Furthermore, moderate heating can induce partial reduction of GO, leading to the removal of some oxygen groups and the restoration of sp^2 carbon pathways. This structural evolution enhances electrical connectivity and reduces interlayer spacing, thereby improving charge transport [5]. Consequently, impedance spectra of GO thin films typically show shrinking semicircles in Nyquist plots with increasing temperature, indicating reduced charge-transfer resistance and improved carrier mobility. The dielectric constant (ε') of GO generally increases with temperature due to enhanced dipolar polarization and space-charge accumulation at interfaces, while dielectric loss (ε'') exhibits relaxation peaks that shift to higher frequencies as relaxation times shorten [6]. These temperature-dependent dielectric features reflect the interplay between dipolar dynamics, interfacial polarization, and thermally activated conduction mechanisms.

Overall, the temperature-dependent electrical and dielectric behavior of graphene oxide thin films arises from a complex combination of structural disorder, interfacial effects, dipole mobility, ionic transport, and thermally assisted hopping conduction. Understanding these mechanisms is essential for optimizing GO-based devices in applications such as flexible electronics, sensors, energy storage systems, and dielectric materials.

2. GRAPHENE OXIDE

2.1 Materials

This study utilized high-purity graphite powder (≥ 99.9 %, Sigma-Aldrich) as the primary precursor material. Other chemicals employed in the synthesis included Potassium Permanganate ($KMnO_4$, Merck), Sulfuric acid (H_2SO_4, Fisher Scientific), Phosphoric acid (H_3PO_4, Sigma-Aldrich), Hydrogen peroxide (H_2O_2, Merck), and N,N-dimethylformamide (DMF, Sigma-Aldrich). Deionized water obtained from a Milli-Q (Merck Millipore) purification system was used throughout the experiments.

2.2 Preparation of Graphene Oxide

Graphite oxide was prepared using a modified Hummers' method [7] a typical procedure, 2 g of high-purity graphite powder was added to a mixture of 80 mL concentrated Sulfuric acid and 20 mL Phosphoric acid in a round-bottom flask. The suspension was continuously stirred at 50 °C to promote the oxidation process. Subsequently, 10 g of Potassium permanganate ($KMnO_4$) was slowly introduced into the reaction mixture while carefully maintaining the temperature below 50 °C to prevent overheating and uncontrolled oxidation. The reaction was allowed to proceed under constant stirring for 12 hours to ensure complete oxidation of graphite. After completion of the reaction, the mixture was cooled naturally to room temperature. Then, 50 mL of hydrogen peroxide (H_2O_2) was gradually added to reduce residual manganese species. The resulting graphite oxide was repeatedly washed with deionized water until the filtrate reached neutral pH. Finally, the product was separated by filtration and dried in a vacuum oven at 60 °C to obtain dry graphite oxide powder.

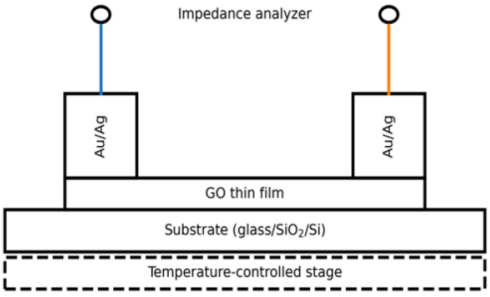


Fig. 1 – Schematic of a GO thin-film impedance measurement

1 g of the prepared graphite oxide was dispersed in 50 mL of DMF using ultrasonication for 30 minutes to achieve a homogeneous suspension. This dispersion was then transferred into a Teflon-lined autoclave and subjected to solvothermal treatment at 180 °C for 12 hours. After cooling to room temperature, the reaction product was collected by filtration and thoroughly washed with deionized water and ethanol to eliminate residual DMF and impurities. The purified graphene oxide was dried in a vacuum oven at 60 °C for 24 hours to remove any remaining moisture and solvents. The dried GO was ultrasonicated in deionized water to obtain a stable colloidal suspension suitable for thin-film fabrication and characterized elsewhere [8].

3. EXPERIMENTAL CONFIGURATION

The electrical properties of graphene oxide (GO) thin films were systematically investigated using impedance spectroscopy under a controlled temperature environment. Measurements were performed using a high-precision LCR meter (HIOKI 3532, Japan). Temperature regulation during the experiments was maintained using a Eurotherm 818p programmable temperature controller with an accuracy of ± 1 K. The temperature range was varied from 300 K to 560 K to examine the temperature-dependent electrical behavior of the GO film. Impedance measurements were recorded over a wide frequency range from 45 Hz to 5 MHz, enabling detailed analysis of the dielectric and electrical response of the film across different time scales. A constant AC oscillation voltage of 1.0 V was applied throughout the measurements to ensure consistency and reproducibility of the data.

Electrical contacts were established using a thin layer of silver paste to ensure good electrode-film interface and stable electrical conduction. This experimental setup provided reliable and accurate characterization of the impedance and electrical properties of the GO film under varying temperature and frequency conditions.

4. THEORETICAL BACKGROUND

4.1 Complex Impedance and Permittivity

The dielectric constant ε' can be expressed as: $\varepsilon' = 1 + \frac{P}{\varepsilon_0 E'}$, where P is the total polarization, E' is the electric field, and ε_0 is the vacuum permittivity [4]. The total polarization is a sum of contributions from electronic (P_e), ionic (P_i), dipolar (P_d), and space-charge polarization (P_s) mechanisms: $P = P_e + P_i + P_d + P_s$. Orientational (dipolar) and space-charge polarizations are particularly temperature sensitive because increased thermal energy facilitates dipole alignment and charge separation at interfaces [4, 5]. In many dielectric materials, the dipolar contribution is often described using Debye relaxation, where ε' depends on the polarization relaxation time τ, $\varepsilon'(\omega) = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1+(\omega\tau)^2}$ and $\tau(T) = \tau_0 \exp\left(\frac{E_a}{k_B T}\right)$. Where, ε_s and ε_∞ are the static and high E_a is the activation energy of polarization, k_B is Boltzmann's constant, and T is absolute temperature.

The electrical response under an alternating electric field can be described using complex impedance formalism. The complex impedance is expressed as: $Z^*(\omega) = Z'(\omega) + jZ''(\omega)$, where $Z'(\omega)$ represents the real part corresponding to resistive processes, and $Z''(\omega)$ is the imaginary part associated with capacitive behavior. Here, $j = \sqrt{(-1)}$ is the imaginary unit. The complex capacitance is related to impedance by: $C^*(\omega) = 1/(j\omega Z^*(\omega))$. From this, the complex permittivity is obtained as: $\varepsilon^*(\omega) = \varepsilon'(\omega) - j\varepsilon''(\omega) = (t/(A\varepsilon_0))\ C^*(\omega)$. where t is the film thickness [9]. A is the electrode area, and ε_0 is the permittivity of free space. The real part ε' represents stored electrical energy, while ε'' corresponds to dielectric loss. The dielectric loss tangent is given by : $\tan\delta = \varepsilon''/\varepsilon'$. A higher value of $\tan\delta$ indicates greater energy dissipation, which is typical for heterogeneous materials such as graphene oxide (GO).

4.2 Relaxation Models

A widely used empirical description of non-Debye relaxation is the Cole-Cole model, written as: $\varepsilon^*(\omega) = \varepsilon_\infty + (\varepsilon_s - \varepsilon_\infty) / [1 + (j\omega\tau)^{(1-\alpha)}]$, where ε_s and ε_∞ are the static and high-frequency permittivities, respectively, τ is the characteristic relaxation time, and $0 \le \alpha < 1$ describes a distribution of relaxation times. For GO, $\alpha > 0$ is expected due to structural disorder, interfacial effects, and heterogeneous conduction pathways.

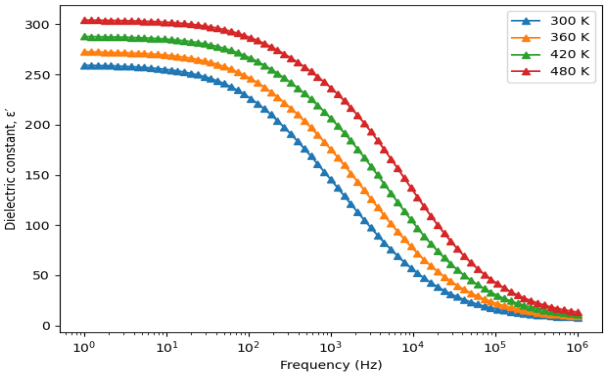


Fig. 2 – Dielectric constant vs frequency

4.3 Conductivity Contributions

In lossy dielectrics, the measured dielectric loss contains both dipolar and conduction components: $\varepsilon''(\omega) = \varepsilon''_{dip}(\omega) + \sigma_{ac}(\omega)/(\varepsilon_0\,\omega)$. The DC conductivity generally follows an Arrhenius temperature dependence: $\sigma_{dc}(T) = \sigma_0 \exp(-E_a/k_B T)$, where E_a is the activation energy and k_B is Boltzmann's constant. For disordered materials such as GO, ac conductivity often follows the Jonscher power law: $\sigma_{ac}(\omega) = \sigma_{dc} + A\ \omega^{s}$ $(0 < s < 1)$ [10]. This behavior suggests hopping conduction between localized states in the GO structure.

5. RESULT AND DISCUSSION

The increase in ε' with temperature in dielectric materials is widely attributed to enhanced dipole mobility and interfacial (Maxwell–Wagner–Sillars) polarization, where charges accumulate at interfaces or grain boundaries when the field oscillates [1, 2, 7]. At low frequencies and higher temperatures the polarization contribution from space charge can be especially significant, resulting in higher ε' values [2, 7].

For graphene oxide materials specifically, recent studies have shown that increasing temperature leads to an increase in ε' due to enhanced charge carrier mobility and thermally activated polarization processes [8, 9]. This trend is consistent with enhanced interfacial polarization at defects and functional groups present in GO.

Temperature generally increases ε′ in the low- and mid-frequency range because thermally activated carriers enhance interfacial polarization (Maxwell–Wagner–Sillars effect) and reduce the effective relaxation time [3]. In practice, the magnitude of this increase depends strongly on film hydration, degree of reduction, and defect chemistry. Low-temperature annealing can also reduce film resistivity while maintaining mechanical stability, as reported for thermally reduced GO under ≤ 300 °C processing.

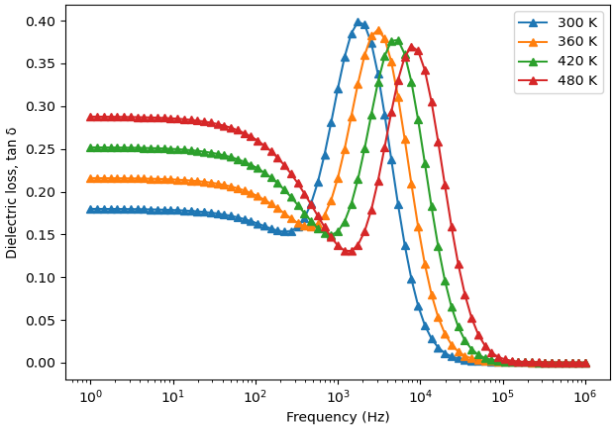


Fig. 3 – The loss tangent (tan δ) versus frequency

Figure 3 shows the loss tangent (tan δ) versus frequency represents the dielectric dissipation behavior of a graphene-oxide thin film under alternating electric fields. At low frequencies, tan δ is high because charge carriers and interfacial dipoles have sufficient time to accumulate at grain boundaries and electrode interfaces, leading to significant energy loss through Maxwell-Wagner polarization and dc conduction. As frequency increases, these slow polarization processes cannot follow the rapidly oscillating field, causing a

systematic decrease in tan δ.

The appearance of a broad relaxation peak indicates distributed dielectric relaxation associated with dipolar reorientation of oxygen functional groups and hopping of localized charge carriers between sp^2 domains. With increasing temperature, the peak shifts toward higher frequency, signifying shorter relaxation time and faster charge dynamics due to thermally activated hopping [11]. Overall, the graph confirms non-Debye relaxation, temperature-enhanced conductivity, and increased dielectric dispersion in GO thin films.

The given Figure 4 is a Nyquist plot ($-Z''$ versus Z') representing the impedance response of a graphene oxide (GO) thin film measured at different temperatures (300-480 K). Nyquist plots are useful for separating bulk, interfacial, and electrode-related contributions to electrical transport.

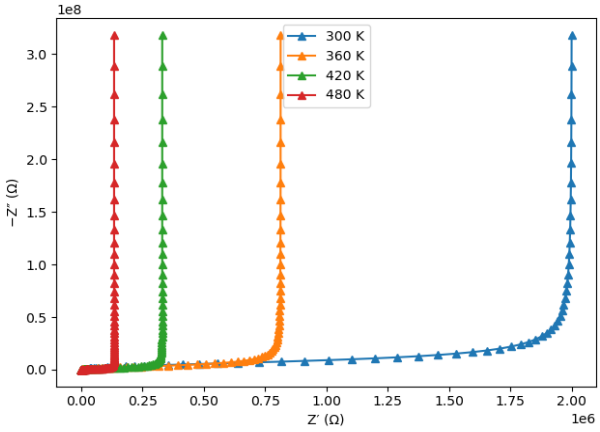


Fig. 4 – Nyquist plot ($-Z''$ versus Z')

At 300 K, the plot exhibits a large intercept on the Z' axis and a highly elongated arc, indicating high resistance and dominant insulating behavior of the GO film. The near-vertical rise of $-Z''$ at low Z' suggests strong capacitive behavior and significant electrode polarization at low frequencies. As the temperature increases to 360 K and 420 K, the intercept on the Z' axis progressively shifts toward lower values, reflecting a reduction in bulk resistance. This behavior arises from thermally activated hopping of charge carriers between localized sp^2 clusters embedded in the insulating sp^3 matrix of graphene oxide. The shrinking of the semicircular feature indicates enhanced conductivity and improved charge transport pathways.

At the highest temperature (480 K), the impedance response becomes dominated by a steep, almost vertical line at very low Z', signifying strong capacitive dominance and minimal resistive contribution. This suggests the onset of semiconductor-like behavior, possibly due to partial thermal reduction of GO and increased carrier density [12]. Overall, the plot demonstrates a clear temperature-driven transition from insulating to more conductive behavior in GO thin films.

6. CONCLUSION

Temperature plays a critical role in governing the dielectric and impedance characteristics of graphene oxide (GO) thin films, making thermal tuning an essential parameter in their functional performance. As temperature increases, the mobility of charge carriers within the GO network is significantly enhanced due to thermally activated hopping between localized sp^2 domains embedded in the insulating sp^3 matrix. This leads to a systematic increase in electrical conductivity and a corresponding reduction in bulk and interfacial resistance. Simultaneously, elevated temperature shortens the characteristic relaxation time of dipolar and space-charge polarization processes, enabling faster dielectric response under alternating electric fields.

In dielectric measurements, higher temperatures typically result in increased low-frequency permittivity due to strengthened Maxwell-Wagner polarization, while relaxation peaks shift toward higher frequencies, confirming accelerated charge dynamics.

Such temperature-dependent behavior is of substantial technological importance. For GO-based sensors, enhanced conductivity at moderate temperatures can improve sensitivity and signal stability. In energy-storage devices such as capacitors, optimized dielectric properties can lead to better charge storage efficiency and reduced energy loss. Moreover, in flexible and wearable electronics, understanding thermal effects ensures reliable performance under varying environmental conditions. Consequently, precise control of temperature-dependent dielectric and impedance properties is crucial for advancing GO-based electronic, sensing, and energy application several thin, layered structures with characteristic wrinkled morphology, while TGA confirms the thermal stability of the GO film, showing significant weight loss primarily associated with oxygenated groups between 200-300 °C. This method advances GO synthesis by offering a scalable approach that maintains control over the structural and functional properties of GO, enhancing its applicability in catalysis, composite materials, and electronic devices. The study underscores the solvothermal method's potential for producing high-quality GO with tunable characteristics, offering valuable insights for future research and applications of graphene-based materials.

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CONFLICTS OF INTEREST

There are no conflicts to declare.

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Вплив температури на діелектричні та імпедансні характеристики тонких плівок оксиду графену

Shib Shankar Biswas[1], Hari Shankar Biswas[1], Dilip Kumar Maiti[2]

[1] *Department of Chemistry, Surendranath College, 700009 Kolkata, India*
[2] *Department of Chemistry, University of Calcutta, University College of Science, Kolkata-09, India*

Тонкі плівки оксиду графену (GO) привернули значну увагу завдяки своїй унікальній комбінації полярних функціональних груп кисню та частково відновленої вуглецевої мережі sp^2, що робить їхню електричну поведінку сильно залежною від температури та частоти. На відміну від чистого графену, GO демонструє гетерогенну структуру, в якій локалізовані носії заряду, структурні дефекти та міжфазні ефекти сильно впливають на його діелектричні та імпедансні характеристики. Ці особливості роблять тонкі плівки GO перспективними для застосування в сенсорах, гнучкій електроніці, пристроях накопичення енергії та системах резистивного перемикання, де температурно-залежні електричні властивості відіграють вирішальну роль. У цій статті представлено практичну основу для розуміння температурно-налаштованої діелектричної спектроскопії та імпедансної поведінки тонких плівок GO, нанесених на ізоляційні підкладки. Аналізуючи комплексні діелектричні властивості, електричний модуль та моделі еквівалентних схем, ми пов'язуємо спостережувану частотну дисперсію діелектричної проникності (ε') та діелектричних втрат (ε'') з мікроскопічними механізмами, такими як міжфазна поляризація Максвелла-Вагнера, стрибки заряду та термічно активований транспорт. Зі збільшенням температури процеси релаксації стають швидшими, зміщуючи піки діелектричних втрат до вищих частот та зменшуючи загальний опір. Діаграми Найквіста демонструють зменшувані півкола при підвищених температурах, що вказує на підвищену рухливість заряду та часткове відновлення GO. Провідність за постійного струму відповідає поведінці типу Арреніуса, що підтверджує термічно активовану провідність.

Ключові слова: Оксид графену, Імпеданс, Провідність, Діелектрична проникність, Рухливість, Електричний модуль, Діелектричні втрати.