

#### Published by NIGERIAN SOCIETY OF PHYSICAL SCIENCES Available online @ https://journaLnsps.org.ng/index.php/jnsps

J. Nig. Soc. Phys. Sci. 7 (2025) 2154

Journal of the Nigerian Society of Physical Sciences

# Improving the thermal stability and dielectric properties of epoxy/phenolic resin type (novolac) composites by incorporating carbon nanofibers (CNFs)

# Elsammani Ali Shokralla\*

Department of Physics, Faculty of Science, Al-Baha University, Saudi Arabia, Aqiq, Saudi Arabia.

# Abstract

This study aims to enhance the thermal stability and dielectric properties of an epoxy/phenolic resin-type novolac blend by incorporating carbon nanofibers (CNFs). The EP/novolac-CNF composite was created by dispersing a 10% weight fraction of CNFs into the EP/novolac blend. Various analyses were conducted to assess the improvements brought by the addition of CNFs. Differential Thermal Analysis (DTA) and Dielectric Measurements (DM) were employed to determine the  $T_g$  values. Thermogravimetric Analysis (TG) and Differential Thermogravimetric Analysis (DTG) were used to evaluate the thermal stability. Results indicated that the inclusion of carbon nanofibers enhanced the thermal stability of the composite, as evidenced by the increased char yield at temperatures exceeding 700°C, reaching 27.08% compared to 8.4% for the EP/novolac blend. Dielectric measurements were conducted across a frequency range of  $10^2 - 10^7$  Hz and a temperature range of 293-463 K. The results revealed a wide dielectric dispersion in all samples, indicating the presence of Debye relaxation and a broad distribution of relaxation times. Eyring's relaxation rate equation was applied to determine the thermodynamic parameters, such as the Gibbs free energy of activation ( $\Delta G$ ) and entropy ( $\Delta S$ ). The results indicated strong intermolecular interactions in all tested samples. The incorporation of carbon nanofibers into the EP/novolac blend led to improvements in thermal stability and dielectric properties. The analysis of various parameters suggests enhanced performance and potential applications of the EP/novolac-CNF composite in relevant fields.

## DOI:10.46481/jnsps.2025.2154

Keywords: Dielectric Constant, Dielectric relaxation, Dissipation factor, Relaxation time, Thermal stability.

Article History : Received: 20 May 2024 Received in revised form: 26 September 2024 Accepted for publication: 15 October 2024 Available online: 23 November 2024

© 2025 The Author(s). Published by the Nigerian Society of Physical Sciences under the terms of the Creative Commons Attribution 4.0 International license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

Communicated by: B. J. Falaye

# 1. Introduction

The development of hybrid polymeric composite materials represents a significant advancement in materials science, offering a wide range of multifunctional properties that meet

\*Corresponding author Tel.: +0-000-000-0000

the demanding requirements of modern technological applications [1–4]. Various composite systems are used in the field of manufacturing technology, including epoxides, phenolics, and epoxy-based composites. The choice of matrix material depends on the specific application requirements, considering factors such as mechanical strength, thermal stability, electrical properties, and chemical resistance. The polymers mentioned—epoxies and phenolics—are selected for their distinct

Email address: sammani970gmail.com (Elsammani Ali Shokralla )

properties, which contribute to the overall performance of the composite material [5, 6].

Nanoparticles possess a significantly larger surface area compared to larger particles, allowing for substantial interaction between the matrix (in this case, the epoxy) and the filler particles (the nanoparticles). Enhanced interaction often leads to improved thermal and dielectric properties, thereby enhancing the performance of the composite material [7–9]. The use of nanoparticles in epoxy-based composites offers several advantages, including improved thermal and dielectric properties, enhanced durability and thermal stability, and reduced weight. These characteristics make nanocomposites desirable for various applications requiring lightweight materials with high performance, such as in aerospace, automotive, and structural engineering industries [10].

Carbon nanofibers (CNFs) possess outstanding thermal conductivity, making them excellent heat conductors. When integrated into composite materials, CNFs can improve thermal management by enhancing heat dissipation and thermal stability. This property is particularly valuable in applications as thermal interface materials (TIMs) in electronic devices, where efficient heat dissipation is critical to performance and reliability [11–13].

In recent years, the development of new TIMs has accelerated through the use of carbon materials such as graphene, carbon nanotubes (CNTs), and carbon nanofibers (CNFs). These materials have been widely applied as fillers in TIMs due to their high thermal conductivity and excellent mechanical properties. Among these carbon materials, carbon nanofibers and carbon nanotubes exhibit highly anisotropic thermal conductivity because of their two-dimensional structure and high aspect ratio. For example, Jeon *et al.* [14] developed cellulosegraphene-based thermal interface papers. According to their studies, although the in-plane thermal conductivity of cellulosegraphene-based thermal interface material paper can reach 7.32,  $W, m^{-1}, K^{-1}$ , the maximum through-plane thermal conductivity is only 0.14,  $W, m^{-1}, K^{-1}$ .

Huang et al. reported the thermal conductivities of current graphene-based polymer nanocomposites. The thermal conductivities of most reported graphene-based polymer nanocomposites were under 10,  $W, m^{-1}, K^{-1}$  due to the high aspect ratio of graphene materials [15]. As for carbon nanotube-based thermal interface materials (TIMs), many studies have demonstrated that the properties of carbon nanotubes cannot be fully exploited because of the presence of defects, high thickness, high interfacial thermal resistance, and low purity [16]. Hong et al. used single-walled carbon nanotubes (SWCNTs) and multiwalled carbon nanotubes (MWCNTs) to fabricate CNT/PMMA (polymethylmethacrylate) composites. The maximum thermal conductivities of SWCNT/PMMA and MWCNT/PMMA composites are 2.43,  $W, m^{-1}, K^{-1}$  and 3.44,  $W, m^{-1}, K^{-1}$ , respectively. They found that the purity and defects of CNTs greatly influence the thermal conductivity of CNT-based TIMs [17].

The aim of this work is to achieve a synergistic enhancement of thermal stability and dielectric properties by leveraging the unique properties of CNFs. The combined effects of CNFs' improved thermal conductivity, dielectric dispersion, and AC



Figure 1. Show schematic illustration of the difference between CNF and CCF.

conductivity contribute to significant improvements in the thermal and dielectric properties of the composite, making it suitable for a wide range of industrial and functional applications.

# 2. Materials and method

The epoxy resins (DGEBA, YD-128) are low molecular weight liquids with epoxide equivalent weight of 176g. The curing agent phthalic anhydride (PA) used in this study was supplied by Kukdo Chem. Co. of Korea. The phenol formalde-hyde type novolac and a curator Hexaminehexa-60 were supplied Bakelite AG company. The Carbon nanofibers (CNF's) purity 99.9, Diameter  $1\mu m$  and length less  $100\mu m$  supplied by Nano shell company, India was used in this study, the Carbon nanofibers (CNF's) specifications shows in Table 1.

#### 3. Samples preparation

The open-mold casting technique is a commonly used method in composite material fabrication, particularly when working with epoxy and novolac blends. In this study, the researcher prepared blends of epoxy and novolac with different weight fractions: specifically, (90% - 10%), (80% - 20%), and (70% - 30%). Among these blends, they selected the (80% - 20%) ratio for further composite preparation. To create the composite, they introduced a fixed weight fraction of carbon nanofibers, specified as 10% by weight. These carbon nanofibers had a diameter of less than  $1 \mu m$  and a length of less than  $100 \mu m$ . The process likely involved mixing the epoxy and

Table 1. Carbon nanofibers (CNF's) specifications.								
Diameter	Length	Purity	Molecular weight	Density	Melting point	Color		
< 1 µm	< 100 µm	99.9%	12.01 g/mol	$2.1 \text{ g/cm}^3$	3925-3970 K	Black		

3





Figure 2. Model stand and sample diagram used for dielectric measurements in the three-electrode method, proposed by Costard *et al.* [18].

novolac components in the desired ratios, followed by the incorporation of carbon nanofibers into the mixture. The open-mold casting technique was then employed to shape and solidify the composite material in a mold, allowing it to take the desired form before curing or setting.

#### 4. Measurements

The dielectric measurement was performed using the threeelectrode method (Guarding electrode method). The specimen was fixed in the specimen holder according to the method of Costard *et al.* [18] and placed in a temperature-controlled oven (Heresies electronic). The high and low terminals of the holder were connected to a dielectric analyzer, Hewlett Packard models HP4274A and HP4275A, while the third terminal was connected to the earth. Three dielectric parameters were measured directly from the above setup: total resistance ( $R_T$ ), total capacitance ( $C_T$ ), and dissipation factor tan( $\delta$ ). Thermal stability was assessed using thermogravimetric analysis (TGA) and differential thermal analysis (DTA) (NETZSCH STA 409PG/PC). Measurements were conducted at a heating rate of 20 K/min over the range of 273-1073 K.

# 5. Results and discussions

#### 5.1. Thermal conductivity

Figure 3 shows the variation of thermal conductivity (k) as a function of temperature for the EP/novolac blend and EP/novolac-CNF's composite. The general behavior of thermal conductivity in all samples shows a linear dependence on

Figure 3. Thermal conductivity of EP/novolac blend and EP/novolac -CNF's composite at the different measurement temperatures.

temperature, also the thermal conductivity of the epoxy is influenced by the presence of the dispersed phase (novolac) and the degree of miscibility within the blend. The behavior observed in the thermal conductivity (k) of EP/novolac blends can be explained by Maxwell's theory, according to Maxwell's theory, the dispersed phase expands the free volume of the EP/novolac blends, thereby reducing the porosity of the network at relatively high temperatures, and the reduction in porosity increases the heat flux and thermal conductivity of the blends [19]. The thermal conductivity of EP/novolac- CNF's composite exhibits a common behavior, increasing linearly with increasing temperature, this behavior is attributed to the increase in intermolecular vibrations as temperature rises, leading to enhanced thermal conductivity [20, 21]. The addition of carbon nanofibers, with their continuous and directional nature, results in higher thermal conductivity values. These findings highlight the importance of material selection and composite design in optimizing thermal conductivity for various applications, including thermal insulation, heat management, and thermal barrier coatings.

# 5.2. DTA analysis

The glass transition temperature is a fundamental property that influences the behavior and performance of polymers across various applications and processing methods. It represents a critical temperature threshold below which polymers experience significant changes in their mechanical and physical properties, making  $T_g$  a key parameter in polymer science, engineering, and material design. The glass transition temperature of the EP/novolac blend and its composite was estimated

Table 2. The glass transition of epoxy, EP/novolac blend and EP/novolac-CNF's

Gropes	Specimens	$T_g(DTA)/K$	$T_g(DM)/K$	Tg(DTA)
Pure EP	EP	361	354	1.95%
EP/novolac	80%EP/20%novolac	378	404	6.78%
EP/novolac -CNF's	EP/novolac-CNF	373,383	404	8.31%



Figure 4. DTA thermograms of EP/novolac blend and EP/novolac - CNF's composite.

using the differential thermal analysis (DTA) technique. Figure 2 shows the differential thermal analysis (DTA) thermograms of the EP/novolac blend and the EP/novolac-CNF composite. Table 2 presents the glass transition temperature ( $T_g$ ) values determined using two different techniques: Differential Thermal Analysis (DTA) and Dielectric Measurements (DM). The EP/novolac blend shows a single  $T_g$  value, but the difference between the DTA and dielectric measurement (DM) values is 6.8, which is relatively large. This difference can be attributed to the fact that dielectric measurements are an approximate method for estimating  $T_g$  and, additionally, to the relatively low compatibility between the EP and novolac networks.

The  $T_g$  value for the EP/novolac-CNF composite was significantly affected by the addition of CNFs. All samples show two  $T_g$  values: the first can be attributed to the polymer blend, and the second may be associated with the interface between the polymers and the reinforcement material [22, 23]. The first  $T_g$  value of the EP/novolac-CNF composite is close to that of the blend ( $\leq$  373 K), while the second  $T_g$  value falls within the range (383  $\leq$   $T_g \leq$  404) K.

## 5.3. TGA analysis

The evaluation of thermal stability using TGA thermograms involves identifying specific point and characteristics in the thermal degradation profile. Here's a breakdown of the parameters commonly used to assess thermal stability. The first step is the onset temperature ( $T_i$ ) represents the temperature at which weight loss or decomposition of the sample begins. Second step is the temperature at 50 weight loss ( $T_{50}$ ), in this temperature the sample loss 50% from the initial weight, T50 provides information about the temperature range over which significant decomposition occurs. The third step is the temperature at which the sample has lost 80% of its initial weight this indicate the temperature at which most of the thermal decomposition has occurred. The last step is the char yield at 1073K0 represent the weight fraction of the residual char or ash remaining after complete thermal degradation of the sample at 1073K.

Based on the description provided, Figures 5(a) illustrate the thermal degradation behavior of EP/novolac blend and EP/novolac- CNF's composite, The TGA thermograms of EP/novolac blend reveal a thermal degradation pattern consisting of four distinct steps, each step corresponds to specific degradation reactions occurring within the material as it is subjected to increasing temperatures. The first step, occurring in the temperature range of 363-393K, is attributed to the loss of moisture and unbound impurities present in the blend. A slight weight loss observed at 548-593K, is explained by the degradation of the propyl chain group in the epoxy resin [17]. The major weight loss, observed in the range of 638-660K, is associated with the loss of the bisphenol-A group present in the blend. Weight loss in the range of 713-853K, is attributed to the breakdown of the methylene linkage group [19]. The final degradation step, occurring at temperatures above 873K, is attributed to thermo-oxidative reactions. Overall, the incorporation of Phenolic resin type novolac into EP resin enhances the thermal stability of the blends, as evidenced by the increase in Tf values. These insights are valuable for optimizing the formulation of EP/novolac blends and designing materials with improved thermal properties for various applications, including structural, automotive, and aerospace industries.

As shown in the TGA spectra Figure 5(b), the thermal degradation behavior of EP/novolac- CNF's composites showing four distinct degradation steps. The first degradation step occurs in the temperature range of 363-408K0, this initial weight loss is attributed to the removal of moisture and other unbound impurities present in the composite material. Typically, at these relatively low temperatures, volatile components such as water and solvent residues are driven off. The second degradation step is observed in the temperature range of 558-613K, this slight weight loss is associated with the degradation of polyol groups present in the composite material. Polyol groups are a common component in epoxy formulations and their decomposition contributes to the overall weight loss at this stage [22–24]. The third degradation step, which results in the major weight loss, occurs over the temperature range of 634-873K, this weight loss is primarily attributed to the degradation of the bisphenol-A group present in the composite material.



Figure 5. TGA and DTG thermograms of EP/novolac blend and EP/novolac -CNF's composite.

Bisphenol-A is a common component found in epoxy resins and its decomposition leads to significant mass loss during this stage of thermal degradation [24]. The fourth degradation step occurs at high temperatures exceeding 1023K, this final weight loss is explained by thermo-oxidative reactions that may occur in the composite material. Thermo-oxidative reactions involve the combination of high temperatures with oxygen, leading to further degradation and decomposition of remaining components in the material [24]. The addition of carbon nanofibers to EP/novolac-CNF's composites improves their thermal stability by increasing char yield and enhancing onset temperatures  $(T_i)$ , temperatures at 50 weight loss  $(T_{50})$ , and potentially extending the thermal degradation range beyond the limits of the TGA analysis. These findings underscore the effectiveness of CNFs as reinforcement materials in enhancing the thermal properties of epoxy based composites, which is crucial for applications requiring high-temperature performance and fire resistance.



Figure 6. Electrical conductivity of EP/novolac blend and EP/novolac -CNF's composite at different frequencies and temperatures.

#### 5.4. Dielectric properties

Figure 6(a) shows the variation of total conductivity  $\sigma_T$ as a function of frequency at various temperatures for the EP/novolac blend and EP/novolac-CNF composites. The results indicate that  $\sigma_T$  at low frequencies, within the range of  $(10^2 - 10^3)$  Hz, is independent of frequency. This behavior is attributed to interfacial polarization, as the DC conductivity dominates at low frequencies. At frequencies greater than 10<sup>3</sup> Hz,  $\sigma_T$  increases rapidly, reaching a value of  $5.04 \times 10^{-7}$  S cm<sup>-1</sup>. This behavior can be associated with the segmental motion of the polymer backbone, which facilitates electronic polarization and the dominance of AC conductivity within this range of frequencies [25].

As shown in Figure 6(b), the EP/novolac-CNF composite exhibits a significant increase in  $\sigma_T$ , rising by three orders of magnitude to reach  $9.11 \times 10^{-4}$  S cm<sup>-1</sup> at 1 MHz. This indicates



Figure 7. The exponent (s) of EP/novolac blend and EP/ Novolac - CNF's Composites at different temperatures.



Figure 8. The activation energy (Eac) of EP/Novolac blend EP/Novolac -CNF's Composites at different frequencies.

a transition to a semiconducting behavior. The substantial increase in  $\sigma_T$  is achieved by incorporating 10 weight percent of carbon nanofibers into the EP/novolac blend, which facilitates the dissipation of static charges. This behavior occurs because all nanofiber molecules in the composite network are interconnected, forming a tri-phase continuity that contributes to charge transport [26].

The values of exponent(s) of EP/novolac blend and EP/novolac-CNF's composite are estimated from the slope of the curves of  $\ln \sigma_{ac}$  versus  $\ln(\omega)$ , are shown in Figure 7, the results reveals that the exponent (s) values are less than unity for all samples. Also these values are decreased with temperature increasing, these values fits with the correlated barrier hopping model (CBH) model [26].



Figure 9. The real part of dielectric constant of EP/novolac blend and EP/novolac -CNF's composite at different frequencies and temperatures.

The activation energies Eac are estimated from the slopes of  $\ln \sigma_{ac}$  versus temperature according to Arrhenius theory. All specimens show three activation energies, and the values of Eac shown in Figures 8 (A & B). The values of activation energies Eac decreases with the frequency increases. This observation indeed supports the discussion regarding the behavior of AC conductivity  $\ln \sigma_{ac}$  with frequency ( $\omega$ ). the discussion suggests that adding of carbon nanofibers into EP/novolac blend, improve the value of Eac values, this improvement is consistent with the behavior of AC conductivity for those composites. The presence of carbon nanofibers is seen as enhancing the overall dielectric behavior of the composite material.

Figures 9(a) and (b) illustrate the variation of the real dielectric constant  $\varepsilon_r$  as a function of frequency ( $\omega$ ) at various temperatures for EP/novolac blend and EP/novolac-CNF's compos-





7

Figure 10. The imaginary part of dielectric constant of EP/novolac blend and EP/novolac -CNF's composite at different frequencies and temperatures.

Figure 11. The dissipation factor of EP/novolac blend and EP/novolac -CNF's composite at different frequencies and temperatures.

ite respectively. The result reveals that values of  $\varepsilon_r$  decreases rapidly with the frequency increases over the range  $10^2 - 10^5$  Hz, while at frequency greater than  $10^5$  Hz the values of  $\varepsilon_r$  are slightly dependent on frequency for all samples. Such behavior is due to the electrode blocking layers' mechanism which is dominated at frequency range over  $10^2 - 10^5$  Hz [27]. Thus, the polarization process exists in this range called the electrode polarization. Also, may be the Maxwell-Wagner polarization is attributed into this range [26–28]. Since in this frequency range two extreme type of conduction behavior of a system formed by discontinuous phases.

Figures 10(a) and (b) shows the variation of the imaginary dielectric constant  $\varepsilon_i$  as a function of frequency ( $\omega$ ) at various temperatures for EP/novolac blend and EP/novolac-CNF's composite respectively. The plots of  $\varepsilon_i$  versus ln( $\omega$ ) shows that the EP/novolac blend and EP/novolac-CNF's composite exhib-

ited a dielectric dispersion over the range  $10^3 - 10^5$  Hz. The observed dielectric dispersion is attributed to ?-relaxation involving cooperative motion of ions with polymer segments. Also, it is appearing that the results demonstrate a consistent shift of loss peaks toward higher frequencies in all tested samples, which includes both the EP/novolac blend and the EP/novolac-CNF's composite. The shift of loss peaks toward higher frequencies indicates a change in the frequency at which maximum energy absorption or dissipation occurs within the material. This phenomenon can be attributed to various factors, such as changes in molecular mobility, interaction between components, or alterations in the relaxation processes within the material citer26.

Figures 11(a) and (b) shows the variation of dissipation factor  $\tan(\delta)$  as a function of frequency at various temperatures for EP/novolac blend and EP/novolac-CNF's composite. The result shows that The loss peaks shift towards higher frequencies with increasing temperature. This phenomenon is attributed



Figure 12. The dielectric parameters ( $\alpha$ ,  $\varepsilon_{\infty}$ ,  $\varepsilon_s$ ,  $\tau_0$ , and  $\tau$ ) of EP/novolac blend and EP/novolac -CNF's composite at different measurements temperatures.

to the strong carrier polarization occurring at the peak zone, which is caused by high densities of low mobility charge carriers citer26,r27. The addition of carbon nanofibers increases the tan( $\delta$ ) values. This indicates that the presence of CNFs enhances the dissipation factor, potentially due to interactions between the Nano fillers and the polymer matrix. The plots of  $tan(\delta)$  versus  $ln(\omega)$  reveal three distinct zones based on frequency ranges. First zone  $(10^2 - 10^3 Hz)$ : In this range, tan( $\delta$ ) decreases as frequency increases. This behavior suggests a certain level of frequency-dependent behavior, possibly related to the relaxation processes within the material. Second zone  $(10^3 - 4 \times 10^4 Hz)$ : Strong dielectric relaxation is observed in this range, marked by the presence of loss peaks. This indicates significant energy dissipation within the material, likely associated with molecular or interfacial relaxation processes. The third zone at high frequencies beyond  $4 \times 10^4$  Hz,  $tan(\delta)$  rapidly decreases as frequency increases. This behavior may indicate a transition to a region where the material exhibits more insulating characteristics, with reduced energy dissipation [27].

Figures 12(a) and (b) shows the values of distribution pa-



Figure 13. The thermodynamic parameters  $(\Delta G, \Delta H, and \Delta S)$  of EP/novolac blend and EP/novolac -CNF's composite at different measurements temperatures.

rameter ( $\alpha$ ), optical dielectric constant ( $\varepsilon_{\infty}$ ), static dielectric constant ( $\varepsilon_s$ ), macroscopic relaxation time ( $\tau$ ), and molecular relaxation time of the EP/novolac blend and EP/novolac-CNF's respectively. The values of  $\alpha$  and  $\tau_0$  systematically decrease with increasing temperature. This trend suggests that the dominant type of polarization in the material is orientation polarization, which is highly sensitive to temperature. As temperature rises, the orientation polarization undergoes slight changes, leading to decreases in  $\alpha$  and  $\tau_0$ . [29]. The values of  $\varepsilon_{\infty}$  for EP/novolac blends lie around 2-3, while for EP/novolac-CNF's composites, they range from 2.45 to 7.34. This indicates variations in the dielectric response of the materials. The range observed for EP/novolac-CNF's composites suggests the presence of different structural configurations or interactions induced by the addition of CNFs. The values of  $\varepsilon_s$  increase with increasing temperature. This behavior can be explained by the activation of dipoles due to thermal energy. As temperature increases, the dipoles gradually become activated and gain freedom of rotation, even though the dielectric remains in the solid phase. This suggests that thermal energy plays a crucial role in activating molecular motions within the material, leading to changes in its dielectric properties [30].

The thermodynamic parameters of EP/novolac blend and EP/novolac- CNF's composites, free energy of activation ( $\Delta G$ ), enthalpy ( $\Delta H$ ), and entropy ( $\Delta S$ ) are estimated from dielectric relaxation measurement, were illustrated in Figure 13(a) and (b). The values of enthalpy are affected by the addition of a small amount of CNF's to the EP/novolac blend. This suggests that the presence of CNF's alters the enthalpic contributions within the material, potentially due to interactions between the polymer matrix and the Nano fillers. Changes in enthalpy can reflect variations in the energy required to maintain the material's structural integrity or undergo phase transitions. The results indicate that the values of  $\Delta G$  and  $\Delta S$  for all tested samples increase with increasing temperature. This observation implies that the activation of segmental dipoles within the material is enhanced as temperature rises, leading to greater energy requirements ( $\Delta G$ ) and increased disorder or randomness ( $\Delta S$ ). This phenomenon is typically attributed to the greater thermal energy available to the molecules, which facilitates their movement and reorientation [31].

## 6. Conclusion

Based on the findings of the study involving the EP/novolac blend and EP/novolac-CNF composites, several key conclusions can be drawn:

- The thermal conductivity of the EP/novolac-CNF composite increases linearly with rising temperature. This behavior is attributed to enhanced intermolecular vibrations at higher temperatures, leading to improved thermal conductivity. The continuous and directional structure of the carbon nanofibers plays a crucial role in enhancing the composite's overall thermal conductivity coefficient.
- Glass transition temperature  $(T_g)$  values were determined using both dielectric measurements (DM) and differential thermal analysis (DTA) techniques. The EP/novolac blend shows a single  $T_g$  value; however, the difference between the DTA and DM values is 6.8 K. For the EP/novolac-CNF composite, the  $T_g$  is significantly affected by the addition of carbon nanofibers. All samples exhibit two  $T_g$  values: the first corresponds to the polymer blend, while the second is associated with the interface between the polymers and the reinforcement material.
- The TGA thermograms of the EP/novolac blend show a thermal degradation pattern consisting of four distinct steps, each linked to specific degradation reactions as the material is subjected to increasing temperatures. The first degradation occurs within the temperature range of 363-393 K, followed by slight weight loss at 548-593 K. The major weight loss occurs between 638-660 K, and weight loss between 713-853 K is attributed to the breakdown of the methylene linkage group. The final degradation step

occurs at temperatures exceeding 873 K. The incorporation of phenolic resin-type novolac into the EP resin enhances the thermal stability of the blend, as evidenced by the increased thermal stability factor ( $T_f$ ) values.

- The addition of carbon nanofibers to the EP/novolac composite improves thermal stability by increasing char yield and raising initial temperatures  $(T_i)$ , the temperature at 50% weight loss  $(T_{50})$ , and potentially extending the thermal degradation range beyond TGA limits, with total degradation reaching up to 1023 K.
- The dielectric properties of the materials, including parameters such as the distribution parameter ( $\alpha$ ), macroscopic relaxation time ( $\tau_0$ ), and static dielectric constant ( $\varepsilon_s$ ), are sensitive to temperature variations. This indicates that temperature significantly influences molecular dynamics and polarization mechanisms within the materials.
- Estimating thermodynamic parameters through dielectric measurements provides valuable insights into the energy dynamics and molecular interactions within the blends and composites. Understanding these parameters is essential for optimizing material compositions and processing conditions, especially for applications requiring high thermal stability and efficient energy management.

# Acknowledgment

I sincerely thank my colleagues at the Department of Physics, Faculty of Science, Al-Baha University, especially Dr. Fakher Al-Waslati and Dr. Ahmed Zakaria, for their valuable assistance.

## References

- G. M. Luz, L. G. Mano, "Mineralized structures in nature: Examples and inspirations for the design of new composite materials and biomaterials", Composites Science and Technology **70** (2010) 1777. http://doi.org/10. 3390/ma6062543.
- [2] K. Song, Y. Zhang, J. Meng, E. Green, N. Tajaddod, H. Li & M. Minus, "Structural polymer-based carbon nanotube composite fibers understanding the processing structure performance relationship", Materials 6 (2013) 2543. http://doi.org/10.3390/ma6062543.
- [3] S. Belloa, J. Agunsoyeb, S. Hassana, M. Zebase, I.Kana & M. Raheem, "Epoxy resin based composites, mechanical and Tribological properties: A review", Tribology in Industry **37** (2015) 500. https://doi.org/10.1007/ 978-981-15-6267-9\_16.
- [4] N. Phong, M. Gabr, L. Anh, V. Duc, A. Betti, K. Okubo, B. Chuong & T. Fujii, "Improved fracture toughness and fatigue life of carbon fiber reinforced epoxy composite due incorporation of rubber nanoparticles", Journal of Material Science 48 (2013) 6039. https://doi.org/10. 1007/s10853-013-7400-z.
- [5] S. Rathinavel, V. Priyadharshini & D. Panda, "A review on carbon nanotube: An overview of synthesis, properties, functionalization, characterization, and the application", Materials Science and Engineering B 268 (2021) 115095. https://doi.org/10.1016/j.mseb.2021.115095.
- [6] M. Kooti. A. Sedeh, K. Gheisari & A. Figuerola, "Synthesis, characterization, and performance of nanocomposites containing reduced graphene oxide, polyaniline, and cobalt ferrite", Physica B: Condensed Matter 612 (2021) 412974. https://doi.org/10.1016/j.physb.2021.412974.

- [7] M. Shettar, C. Kowshik, M. Manjunath & P. Hiremath, "Experimental investigation on mechanical and wear properties of nanoclay epoxy composites", Journal of Materials Research and Technology 9 (2020) 9108. https://doi.org/10.1016/j.jmrt.2020.06.058.
- [8] M. Jawaid, S. Chee, M. Asim, N. Saba & S. Kalia, "Sustainable kenaf/bamboo fibers/clay hybrid nanocomposites: Properties, environmental aspects and applications", Journal of Cleaner Production 330 (2022) 129938. https://doi.org/10.1016/j.jclepro.2021.12993.
- [9] S. Moharana & B. Sahu, Synthesis and properties of epoxy-based composites, IntechOpen Limited, London, United Kingdom, (2022) pp. 1-28. https://doi.org/10.5772/intechopen.104119.
- [10] Y. Baghdadi, L. Youssef, K. Bouhadir, M. Harb, S. Mustapha, D. Patra & A. Riza, "Thermal and mechanical properties of epoxy resin reinforced with modified iron oxide nanoparticles", Journal of Applied Polymer Science 138 (2021) 50533. https://doi.org/10.1002/app.49330.
- [11] M. Seong & D. Kim, "Effects of facile amine-functionalization on the physical properties of epoxy/graphene Nano platelets nanocomposites", Journal of Applied Polymer Science 132 (2015) 42269.https://doi.org/10. 1002/app.42269.
- [12] D. Yadav, F. Amini & A. Ehrmann, "Recent advances in carbon nanofibers and their applications: a review", European Polymer Journal 138 (2020) 109963. https://doi.org/10.1016/j.eurpolymj.2020.109963.
- [13] X. Yang, Y. Chen, C. Zhang, G. Duan & S. Jiang, "Electrospun carbon nanofibers and their reinforced composites: Preparation, modification, applications, and perspectives", Composites Part B Engineering 249 (2023) 110386. https://doi.org/10.1016/j.compositesb.2022.110386.
- [14] M. Hao, X. Qian, Y. Zhang, J. Yang, C. Li, H. Gong, X. Wang, P. Wang, L. Liu & Y. Huang, "Thermal conductivity enhancement of carbon fiber/epoxy composites via constructing three-dimensionally aligned hybrid thermal conductive structures on fiber surfaces", Composites Science and Technology 231 (2023) 109800.https://doi.org/10.1016/j. compscitech.2022.109800.
- [15] D. Jeon, S. H. Kim, W. Choi & C. Byon, "An experimental study on the thermal performance of cellulose-graphene-based thermal interface materials", International Journal Heat Mass Transfer 132 (2019) 944. https://doi.org/10.14447/jnmes.v17i2.433.
- [16] J. Khan., S. Momin, & M. Mariatti, "Review on advanced carbon-based thermal interface materials for electronic devices", Carbon 168 (2020) 65. https://doi.org/10.1016/j.carbon.2020.06.012.
- [17] N. Greef, L. Zhang, A. Magrez, L. Forro, J. Locquet, L. Verpoest & J. Seo, "Direct growth of carbon nanotubes on carbon fibers: Effect of the CVD parameters on the degradation of mechanical properties of carbon fibers", Diamond Related Mater **51** (2015) 39. https://doi.org/10.1016/j. diamond.2014.11.002
- [18] J. Costard, M. Ender, M. Weiss, & E. Tiffee, "Three-electrode setups for lithium-ion batteries II. experimental study of different reference electrode designs and their implications for half-cell impedance spectra", Journal of The Electrochemical Society 164 (2017) 80. http://doi.org/10. 1149/2.0241702jes.
- [19] W. Hong & N. Tai, "Investigations on the thermal conductivity of composite reinforced with carbon nanotubes". Diamond Related Matter 17 (2008) 1577. https://doi.org/10.1016/j.diamond.2008.03.037.

- [20] A. Shimamura, Y. Hotta, H.i Hyuga, M. Hotta & K. Hirao, "Improving the thermal conductivity of epoxy composites using a combustionsynthesized -Si3N4 filler with randomly oriented grains", Scientific Reports 10 (2020) 14926.https://doi.org/10.1038/s41598-020-71745-w.
- [21] M. Haruki, "Thermal Conductivity for Polymer Composite Materials", Journal of Chemical Engineering of Japan 54 (2021) 186. https://doi.org/ 10.1252/jcej.20we136.
- [22] S. A. Shokralla & N. S. Al-Muaikel, "Thermal properties of Epoxy (DGEBA) /Phenolic resin (Novolac) Blends", The Arabian Journal for Science and Engineering B 35 (2010) 7. https://www.researchgate.net/profile/Nayef-Al-Muaikel/publication/ 267564344\_Thermal\_properties\_of\_epoxy\_DGEBAphenolic\_ resin\_NOVOLAC\_blends/links/5517bc820cf2f7d80a3b8fa6/ Thermal-properties-of-epoxy-DGEBA-phenolic-resin-NOVOLAC-blends. pdf.
- [23] M.Lila, G.Saini, M. Kannan & I. Singh, "Effect of Fiber Type on Thermal and Mechanical Behavior of Epoxy Based Composites", Fibers and Polymers 18 (2017) 806. https://doi.org/10.1007/s12221-017-1147-0.
- [24] V. Chinnasamy, S.Subramani, S. Palaniappan, B. Mylsamy & K. Aruchamy, "Characterization on thermal properties of glass fiber and kevlar fiber with modified epoxy hybrid composites", Journal of Mater Research Technology 9 (2020) 3158.https://doi.org/10.1016/j.jmrt.2020. 01.061.
- [25] J. Zhang, M. Ma, Y. Bi , Z. Liao, Y. Ma, W. Huang, P. Lyu & C. Feng, "A review of epoxy-based composite materials: Synthesis, structure and application for electromagnetic wave absorption", Journal of Alloys and Compounds **922** 20 (2022) 166096. https://doi.org/10.1016/j.jallcom. 2022.166096.
- [26] E. A. Shokralla, "Dielectric relaxation, electric conductivity and thermodynamic studies on epoxy polyurethane blend and their composites", International journal of Materials Science and Applications 13 (2024) 6.https://doi.org/10.11648/ijmsa.20241301.12.
- [27] Z. Wang, M. Yang, Y. Cheng, J. Liu, G. Wu & H. Wu, "Dielectric properties and thermal conductivity of epoxy composites using quantum-sized silver decorated core/shell structured alumina/polydopamine", Composites Part A: Applied Science and Manufacturing **118** (2019) 302. https: //doi.org/10.1016/j.compositesa.2018.12.022.
- [28] Z. M. Elimat, M. S. Hamideen, K. I. Schulte, H. Wittich, A. de la Vega, M. Wichmann & S. Buschhorn, "Dielectric properties of epoxy/short carbon fiber composites", Journal of Materials Science 45 (2010) 5196.https:// doi.org/10.1007/s10853-010-4557-6.
- [29] A. A. Kareem, "Enhanced thermal and electrical properties of epoxy/carbon fiber silicon carbide composites", Advanced Composites Letter 19 (2020) 1. https://doi.org/10.1177/2633366X1989459.
- [30] H. Prashanth, P. Shanmugasundram, E. Jayamani & K. Heng, "A comprehensive review on dielectric composites: Classification of dielectric composites", Renewable and Sustainable Energy Reviews 157 (2022) 112075. https://doi.org/10.1016/j.rser.2022.112075.
- [31] V. Kumar, S. Ramakrishna, S. Rajendran & S. Surendran, "Enhancing the material properties of carbon fiber epoxy composite by incorporating electrospun polyacrylonitrile nanofibers", Materials Today Proceedings 67 (2022) 1. https://doi.org/10.1016/j.matpr.2022.04.818.

10