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Effects of Carbon Nanotubes on Thermal Behavior of Epoxy Resin Composites

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Abstract: Human society's need to build low-weight, high-strength and durable structures has increased the demand for composite materials. In this case, composites are used where high mechanical strength, low weight, sound and thermal insulation properties are required. One of the most important issues now is designing materials and coatings aimed at reducing heat loss and resisting high temperatures. One way to address this problem is to develop a technique for preparing and applying composite materials that slow down their heating applied to a surface. In this study, carbon nanotubes (CNTs) reinforced composites were fabricated using silicone molding to be applied to honeycomb sandwich structures. To determine the effect of CNTs on the thermal behavior of the sandwich panels, different weight percentages of this material (0.025, 0.05. 0.075 wt.%) were added to the epoxy resin. The results showed that the thermal stability of the epoxy composites was directly related to the increase in the percentage of CNTs as the CNT content increased to 0.075 wt.%, and the thermal degradation temperature of the epoxy composites increased by 14 °C. In addition, the energy absorption increased by 4.6% with an increase in CNTs up to 0.075 wt.%. Density measurements showed that the density of the nanocomposite samples increased by adding CNTs to pure epoxy resin. The actual densities of the samples reinforced with 0.025, 0.05, and 0.075 wt.% CNTs are 0.925, 0.926, and 0.927 of the theoretical density, respectively. Since the CNT dispersion uniformity in the epoxy matrix can significantly affect the properties of the composites, in this study, a new method of dispersing CNTs in the epoxy resin matrix resulted in higher thermal conductivity while using lower amounts of CNTs compared to other studies. The storage modulus of the epoxy matrix composites reinforced with 0.05 wt.% in this study was 25.9% and 6.9% higher than that from the previous study reinforced with 0.1 wt.% and 0.25 wt.% CNTs, respectively. Furthermore, the tanδ and loss modulus of the composite reinforced with 0.05 wt.% CNTs in this study were 52% and 54.5% higher than that from the previous study with 0.1 wt.% CNTs, respectively. This study provided an optimal approach for designers and engineers who want to effectively design their composite honeycomb sandwich structure with better thermal properties.

Keywords: epoxy composites; carbon nanotubes (CNTs); thermal stability; energy absorption; density; honeycomb sandwich structure; CNT dispersion

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1. Introduction

Polymer-based composites are widely used in industries such as marine, civil, aerospace, biomedical, and power industries, to name a few. Polymer sandwich panels are used in applications where lightweight structures with high strength and thermal and sound insulation are needed. As a result, their applications have extended to the automotive and construction industries. The use of sandwich panels as structural elements is not limited to the vehicles, docks, and reconstruction industries or as a replacement for concrete bridges; they are also used as main elements in roofs and dividing walls [1–3]. CNT-reinforced epoxy honeycomb sandwich panels have a wide range of applications due to their special mechanical and thermal properties. Their mechanical properties, such as high rigidity and strength with low weight, make them suitable for architectural, aerospace, and military applications [4]. Their high thermal conductivity makes them important for applications such as electronics that require high thermal conductivity. Therefore, honeycomb sandwich structures made of CNT-reinforced epoxy resin matrix can be used in applications that require high flexural, strength, and thermal conductivity simultaneously, such as reusable launch vehicles based on load-carrying structures [5] and ablative heat shields [6] that require high strength and thermal conductivity.

In fact, sandwich panels are special types of multilayer composites. In this regard, a sandwich structure consists of a lightweight, thick core covered by two high-strength thin shells. The shells are usually very hard, while the inner core is relatively weak and flexible. When combining them into a sandwich panel, a highly lightweight and rigid structure has been built [7].

Epoxy resins are used in many applications, such as floor coverings, glues, coatings, tool making and composites. Epoxy resins are widely used in composites and provide desirable properties such as desirable strength and hardness, thermal stability, and strength against chemical environments. Recently, researchers have started to study epoxy composites containing nanoparticles to analyze their physical, mechanical, and chemical properties. In this context, the desirable properties obtained due to the addition of nanoparticles to epoxy resins increase the tendency to use nanoparticles. Among many nano-reinforcements used in the manufacture of nanocomposites, carbon nanotubes (CNTs) play an important role in improving the physical and mechanical properties of epoxy resins, considering their unique properties such as low density, high thermal conductivity, and high strength and hardness. Currently, composite materials used for thermal fillers do not provide any electrical properties. CNTs have excellent mechanical, thermal, and electrical properties. CNTs have a strong crystal lattice, giving them good physical and thermal properties [8,9].

It can be observed from the literature that CNTs possess high electrical and thermal conductivity as well as excellent mechanical properties [10]. CNTs are basically one-dimensional graphene structures rolled into a cylindrical shape. So, their geometric properties, which is the aspect ratio, can affect the overall properties of CNT-reinforced polymers. Furthermore, their chirality affects their thermal conductivity. Studies have shown that the effect of chirality on the conductivity of short-length CNTs is greater than that of long-length CNTs, and the larger the chiral angle, the greater the conductivity of CNTs. In addition, zigzag CNTs are less thermally and electrically conductive than armchair CNTs. Therefore, as the tube aspect ratio becomes larger, the conductivity increases while the effect of chirality on the conductivity decreases [11]. Moreover, another factor that can contribute to the increment of thermal properties is homogenous dispersion [12]. The way the CNTs are dispersed in the epoxy can significantly affect the properties of the composites. Loos et al. [13] and Caglayan et al. [14] studied the storage modulus of the epoxy resin matrix composites reinforced with 0.25 wt.% and 0.75 wt.% CNTs, respectively. Her and Lin [15] studied the tanδ and loss modulus of epoxy composites reinforced with 0.1 wt.% CNTs. The dispersion method can be improved so as the performance of the composites.

Heat conduction is based on the principle that the vibration of the atomic lattice is transmitted to another lattice next to it and vibrates; since CNTs have a regular crystalline structure, vibration is not scattered but quickly transmitted from one end to the other [16]. Long-CNTs can significantly improve thermal conductivity and diffusivity compared with short ones [17]. It is predicted that the theoretical thermal conductivity of single-walled carbon nanotubes (SWNTs) can reach more than 6600 W/mK [18]. Experimental measurements on individual multi-walled carbon nanotubes (MWNTs) and SWNTs have shown high thermal conductivity of over 3000 W/mK [19] and 3500 W/mK [20], respectively. Wang and Qiu [21] studied the effect of CNTs on the thermal conductivity of glass fiber/polymer composites. They found that the thermal conductivity of the polymer reinforced with 3 wt.% of MWNTs was 1.5 times that of the pure glass fiber/polymer composite. Research has shown that if the amount of CNTs' weight percentage is more than 0.1 wt.%, it cannot significantly increase the thermal conductivity of the composites. For example, with the addition of 0.1 wt.% and 0.25 wt.% of SWNTs, the storage modulus of the composite increased by 9% and 16%, respectively [13]. For 0.25 wt.% and 0.75 wt.% of MWNT-reinforced epoxy composites, the storage modulus of the composites can be increased by 28% and 35%, respectively [14]. Dispersing CNTs in polymers can slightly increase the thermal conductivity of nanotube–polymer composites compared to pristine polymers, while the thermal behavior of CNT composites largely depends on the processing conditions [15,22–24].

Most of the research has been conducted on aluminum honeycomb sandwich panels. Therefore, most sandwich panels are metallic at present. The main goal of this study is to fabricate CNT-reinforced epoxy matrix honeycomb polymer sandwich panels. Honeycomb sandwich panels made of epoxy polymer and reinforced by CNTs have not been found in research. There are mainly two types of research on honeycomb-reinforced composites. One was made of metal honeycombs, and the other was epoxy reinforced with CNTs. Therefore, this research investigates the thermal properties of CNT-reinforced epoxy honeycomb sandwich panels, which have not been studied by other researchers. Loos et al. [13] and Caglayan et al. [14] studied the effect of CNTs on the thermal conductivity of epoxy matrix composites, Cetin [25] studied the thermal conductivity of MWCNTs reinforced polyurethane aluminum honeycomb sandwich panels. CNT-reinforced epoxy composite is suitable for devices increasing thermal conductivity, such as thermal storage devices and heat exchangers. Moreover, they are suited for high-strength structures needed for lightweight applications such as aerospace, marine, automotive, and sports [26,27]. Moreover, a new method of dispersing CNTs in epoxy resin was applied, which can significantly affect the thermal conductivity of the polymer composite. An attempt was made to reinforce epoxy resins with different weight percentages of CNTs (0.0 wt.%, 0.025 wt.%, 0.050 wt.%, and 0.075 wt.%) based on the previous research outcomes and the effects of these additives on the thermal behavior of polymer composites was evaluated.

2. Materials and Methods

To fabricate standard epoxy composites with/without reinforcement, the following steps were taken.

Standard samples were made of plexiglass and silicone castings.

2.1. Materials

In this work, bisphenol A-based epoxy resin was used. This resin exhibits strong chemical performance over a broad spectrum of acidic, oxidative alkaline environments, ambient and higher temperatures [28]. Moreover, cycloaliphatic amin was used as a hardener in a ratio of 1:4, and the curing time of the resin at room temperature was measured for 120 min.

A silicone mold was used for epoxy molding. The silicone used was of RTV type. This material has two components, one of which is resin, and the other is hardener. The mixing ratio of silicone used in this research is 5 wt.%, which means that the amount of hardener is equal to 5 wt.% of the resin. After mixing these two components at room temperature for 180 min, the shape changed to become an elastomer (a flexible polymer).

To strengthen the epoxy resin, carboxyl-functionalized MWNTs (diameter: 20–30 nm) were used as reinforcement materials. Table 1 shows the properties of the CNTs used in this experiment.

Table 1. The properties of the used CNTs.

Diameter	20–30 nm
Length	30 μm
Functional Group	Carboxyl (COOH)
Purity	>95 wt.%
Impurity	<1.5 wt.%

2.2. Molding Manufacturing

2.2.1. Making Plexiglass Molds

Plexiglass molds suitable for silicone molding were made. In this way, the shape of the honeycomb plexiglass mold was first designed by AutoCAD software, and then the shape was cut on the plexiglass with a laser cutting machine. So, the plexiglass mold was prepared for silicone loading.

2.2.2. Making Silicone Molds

In order to produce the silicone mold, the wall of the plexiglass mold was first coated with grease so that the silicone could be separated from the mold after curing. Then, the silicone resin and the hardener were mixed at a ratio of 5% by weight and mixed thoroughly. The resulting mixture was poured into the plexiglass mold, and after 3 h, the silicone mold was completely dry. The reason for using the silicone mold to load epoxy is the flexibility of silicone; since epoxy is inflexible, a flexible mold should be used so that it can be removed from the mold after curing, whereas if using a non-flexible mold, such as plexiglass, it is impossible to separate the epoxy from it.

2.3. Fabrication of Standard Samples

2.3.1. Pure Epoxy Resin

To cast the samples, first, the walls of the rectangular silicone mold with dimensions 30 mm× 10 mm × 2.5 mm were lubricated with grease. Then, the epoxy resin and curing agent were mixed in a ratio of 1:4 and stirred. The mixture was cast as required and placed under vacuum at 80 Torr for 60 min. After 120 min, the curing process was completed, and the pure rectangular epoxy was extracted from the casting mode.

2.3.2. CNT Reinforced Epoxy

The reinforced honeycomb core was made of different weight percentages of CNTs (0.025, 0.05, and 0.075 wt.%) since, according to the research, in the case of higher CNT content, the CNTs tend to agglomerate and behave like inclusion defects, resulting in a decrease rather than an increase in properties of the composites. Since the performance of polymer nanocomposites depends highly on the dispersion of nano-reinforcements in the substrate and the compatibility of polymer fillers and surface interactions, proper distribution of CNTs in epoxy resin is essential [29]. Next, the CNT dispersion will be described.

First, weigh a certain amount of epoxy resin (relative to the casting size) and then weigh the CNTs at 0.025 wt.%, 0.050 wt.%, and 0.075 wt.% of the epoxy weight. Add a certain amount of acetone into the CNTs and put them into the probe ultrasonic device (70 W, cycle of 10 s) for 30 min; the device is made by IKA, a company in Staufen, Germany. This is to eliminate the clot state of the CNTs. Then, add this suspension to the epoxy resin, and stir the mixture at a temperature of 40 °C with a magnetic stirrer for 1 h, thereby completely extracting acetone. The magnetic hotplate stirrer is made in MTOPS in Seoul, South Korea. Finally, add the hardener to the homogeneous solution of epoxy resin and CNTs and stir well [24]. Pour the solution into a pre-lubricated silicone cast and place the casting in a vacuum at a pressure of 80 Torr for 60 min to extract air bubbles from the casting. After 120 min, curing will be complete, remove the samples from the casting.

The vacuum machine is made by KNF, a company in Hamburg, Germany. Figure 1 is a schematic diagram of the steps for fabricating the reinforced samples.

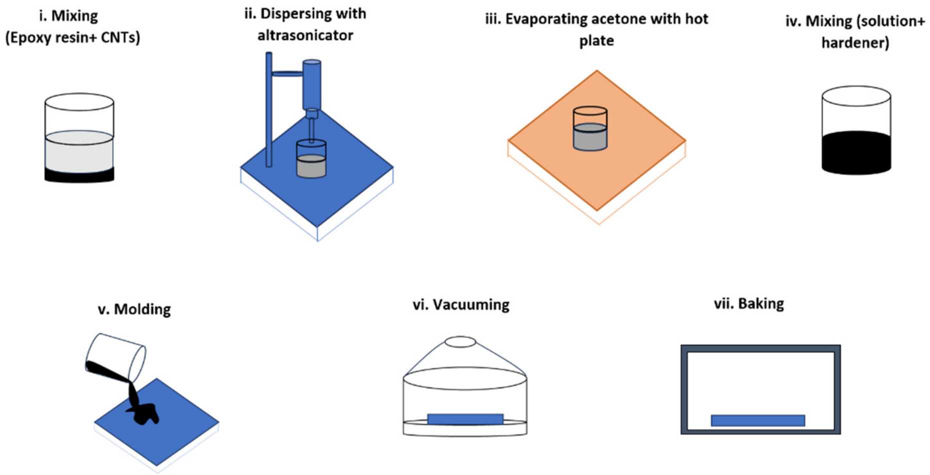

Figure 1. A schematic of the steps for fabricating the reinforced samples.

The purpose of defoaming is to remove air bubbles in the epoxy/CNT solution. The vacuum device works by applying opposing pressure to the material and then removing that pressure. This technique causes air bubbles to move from the solution to the surface, where they are easily removed from the solution when they reach the surface [30]. Figure 2 shows the CNT-reinforced epoxy matrix honeycomb composite sample.

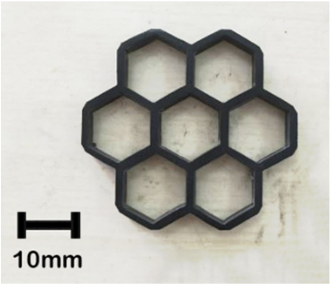

Figure 2. CNT-reinforced epoxy honeycomb composite sample.

Figure 3 shows the microscopic images of the samples of the pure epoxy resin and CNT-reinforced epoxy composites, which shows that the CNTs can be well dispersed in the resin.

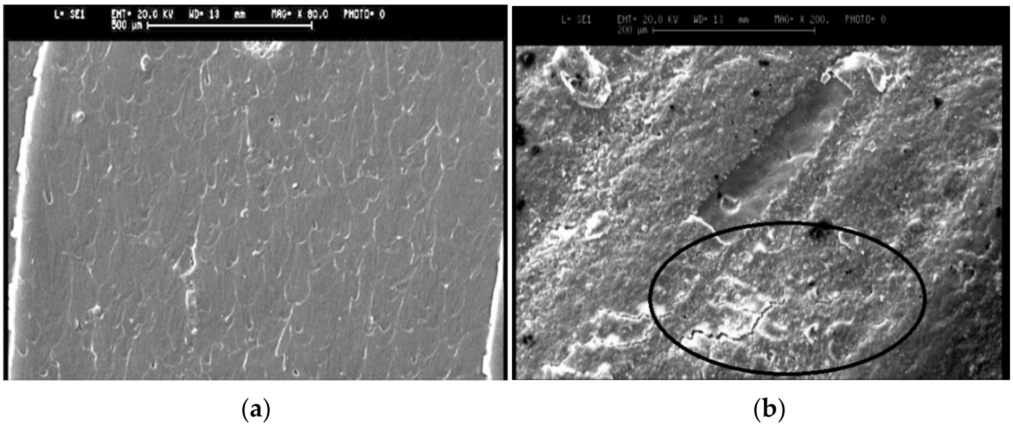

Figure 3. Microscopic images of (**a**) pure epoxy, (**b**) CNT-reinforced epoxy composites (CNTs have been circled out).

2.4. Dynamic Mechanical Thermal Analysis (DMTA)

DMTA analysis is one of the thermal analysis methods widely used to study and identify polymers. This analysis was performed to study the viscoelasticity of epoxy resins in their pure state and reinforced with CNTs [31]. Mechanical changes in polymer sample behavior were measured in terms of temperature, time, frequency, stress, and strain. Samples were analyzed by a DMTA-Triton device, Triton, model 2000 and made in the UK. DMTA analysis was performed on a rectangular sample with a length of 30 mm, a width of 10 mm and a thickness of 2.5 mm, a frequency of 1 Hz, a temperature range of 20 to 200 °C, a heating rate of 5 °C/min and in bending mode. In this work, the storage modulus, loss modulus, and tanδ were investigated.

2.5. Thermogravimetric Analysis (TGA)

TGA was used to study the thermal properties of epoxy resins and to investigate the effect of the addition of CNTs on these properties. This analysis is widely used to study the thermal stability of the polymer as a function of temperature [32]. The role of temperature in evaluating the degradability of epoxy resins was studied by TGA. In this test, the weight of the testing material was continuously measured and recorded as the temperature increased. Based on this analysis, the degradability rate of epoxy resin and epoxy reinforced with different percentages of CNTs can be obtained. The tests were performed on 6 mg samples with an SDT Q600 V20.9 Build 20 device made by TA Instruments, a company based in the USA, at a heating rate of 10 °C/min and under N_2 atmosphere in the temperature range of 25 to 600 °C.

3. Results and Discussion

3.1. Dynamic Mechanical Thermal Analysis (DMTA)

The storage modulus (E') and loss modulus (E'') in viscoelastic materials measure the energy stored (representing the elastic part) and the energy dissipated as heat (representing the viscous part), respectively. The tensile storage and loss moduli are defined as follows [33]:

$$\text{Storage}: \; E' = \frac{\sigma_0}{\varepsilon_0}\cos\delta \tag{1}$$

$$\text{Loss}: \; E'' = \frac{\sigma_0}{\varepsilon_0}\sin\delta \tag{2}$$

where δ is the phase lag between stress and strain. Stress and strain in a viscoelastic material can be expressed by the following expressions [33]:

$$\text{Stress}: \; \sigma = \sigma_0 \sin(\omega t + \delta) \tag{3}$$

$$\text{Strain}: \; \varepsilon = \varepsilon_0 \sin(\omega t) \tag{4}$$

where t is the time, ωt is the phase angle change, ε_0 and σ_0 are the initial (t = 0) strain and stress, respectively.

The ratio of the loss modulus to the storage modulus for a viscoelastic material used in tensile testing is defined as [33]

$$\tan\delta = \frac{E''}{E'} \tag{5}$$

It is often called damping, and it is a measure of the energy dissipation of a material. Damping is the dissipation of energy in a material under cyclic load. It measures how well a material sheds energy and is reported as the tangent of the phase angle. It tells us how well a material absorbs energy. For materials with tanδ greater than 1, the energy-dissipative viscous component of the modulus dominates [34].

The storage modulus plot of the pure and nanocomposite samples is shown in Figure 4. The storage modulus values of all nano-composite samples are higher than that of the pure resin samples. The E' values increase with the addition of CNTs, which is due to the limitation of the polymer chain movement only by the addition of CNTs [35]. Thus, the epoxy was hardened below the T_g (glass transition temperature) point, and as a result, the range of the storage modulus increased. Due to the flexibility of the polymer chains, the value dropped significantly after the T_g point. The highest storage modulus values were obtained for the samples reinforced with 0.075 wt.% CNTs due to the presence of a strong interface, which effectively transmits force between the epoxy and the CNTs. This result indicates that these nanocomposite samples have the best elastic portion energy absorption behavior. In addition, the T_g firstly slightly increases from around 65–70 °C for the pure epoxy sample to around 70 °C for the nanocomposite sample reinforced with 0.025 wt.% CNTs, then the T_g dramatically decreases to around 42–45 °C for the nanocomposite sample reinforced by 0.05 wt.% CNTs. Thereafter, the T_g stays stable around 40–43 °C for the nanocomposite sample reinforced with 0.075 wt.% CNTs, indicating a threshold existing for the optimal effect of the CNT content on the material behavior.

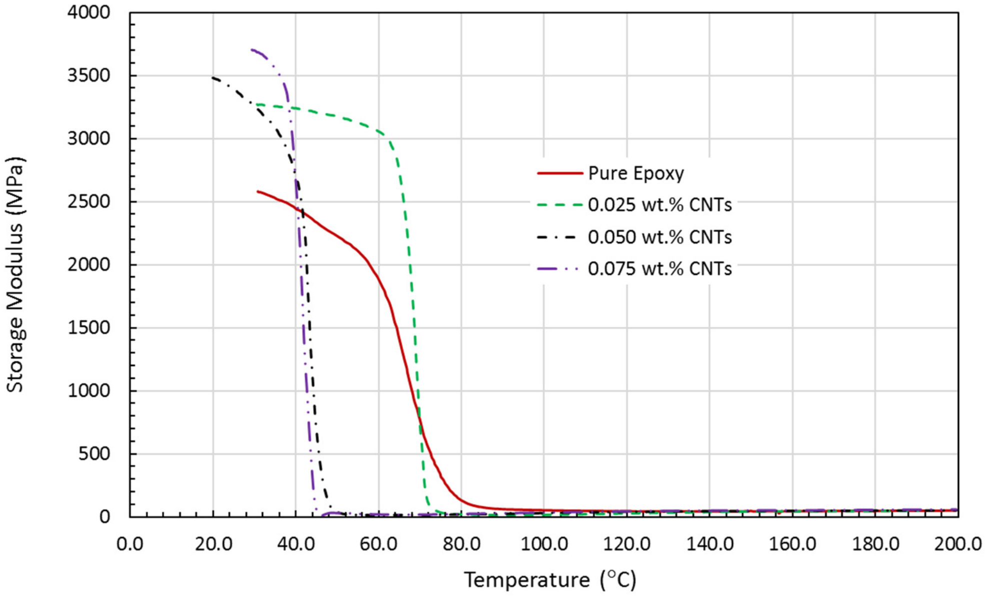

Figure 4. Storage modulus vs. temperatures for pure resin and nanocomposite samples.

According to Figure 5, the maximum storage modulus of the epoxy sample reinforced with 0.075 wt.% CNTs increased by 43.8% compared to pure epoxy resin samples. The storage modulus of the samples reinforced with 0.025 and 0.05 wt.% of CNTs increased by 26.8% and 34.9%, respectively.

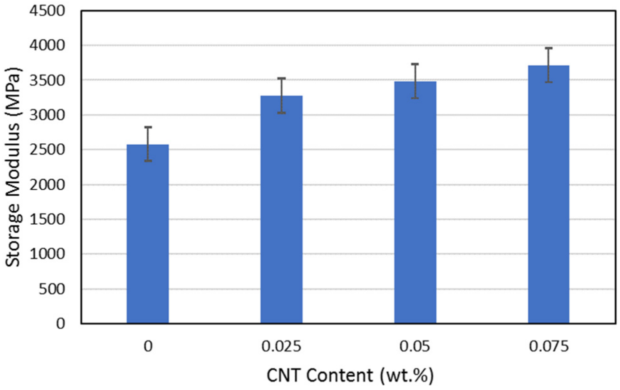

Figure 5. Comparison of the storage modulus of samples vs. CNT wt.%.

As shown in Figure 6, the loss modulus (E'') of the nano-composite samples increased compared to the pure resin samples. The loss modulus also follows the same trend as that observed in the storage modulus, which indicates a greater amount of energy loss in the nanocomposite samples compared to the pure epoxy sample. However, the temperatures corresponding to the maximum loss modulus dramatically increase from 41.4 °C to 64.7 °C with CNT contents increase from 0 wt.% to 0.025 wt.%, then slightly increase from 64.7 °C to 70.6 °C with CNT contents increase from 0.025 wt.% to 0.050 wt.%. Finally, the temperatures start dramatically decreasing from 70.6 °C to 43.2 °C. This phenomenon reflects again the effect of the CNT content on the material behavior and indicates a threshold existing for the CNT content to achieve the optimal effect.

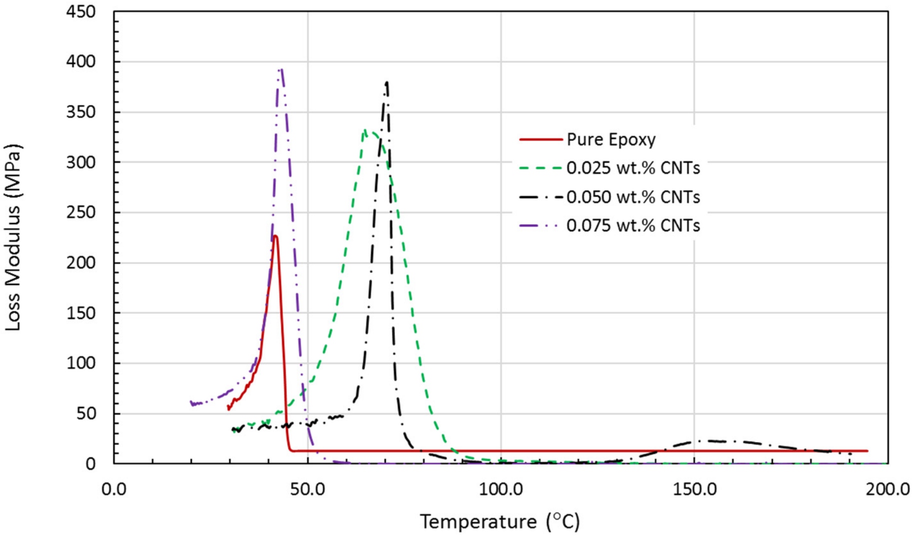

Figure 6. Loss modulus vs. temperatures for pure and nanocomposite samples.

According to Figure 7, the loss modulus of the nano-composite sample reinforced with 0.075 wt.% CNTs increased by 74.4% compared to pure epoxy resin. Furthermore, the loss moduli of the samples reinforced with 0.025 and 0.05 wt.% CNTs increased by 48.8% and 67.0%, respectively, compared to the pure epoxy samples.

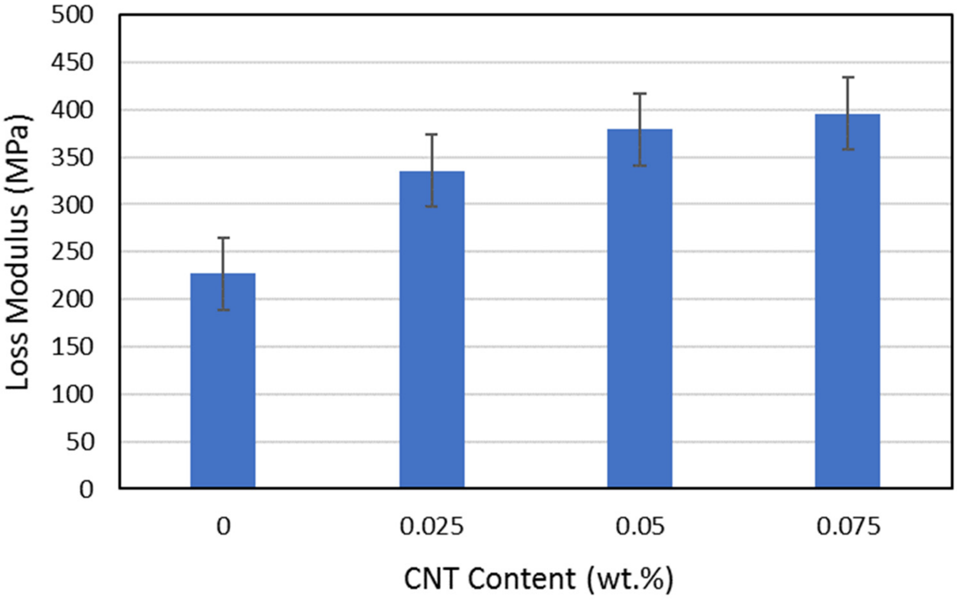

Figure 7. Comparison of the loss modulus of samples with different wt.% of CNTs.

Figure 8 shows the tanδ for the pure epoxy and nano-composite samples vs. temperature. As wt.% CNTs increases, the tanδ increases. The tanδ also follows the trend of the results observed in the storage and loss moduli, which indicates larger tanδ values for the nanocomposite samples compared to the pure samples. The tanδ values of the nanocomposite samples are all greater than one, indicating that the energy-dissipative viscous components of the modulus of these samples dominate, especially for the nanocomposite sample with 0.075 wt.% CNTs, the tanδ reaches 5.138. In addition, the temperatures corresponding to the maximum tanδ change with the CNT content in a similar pattern as the above material behavior.

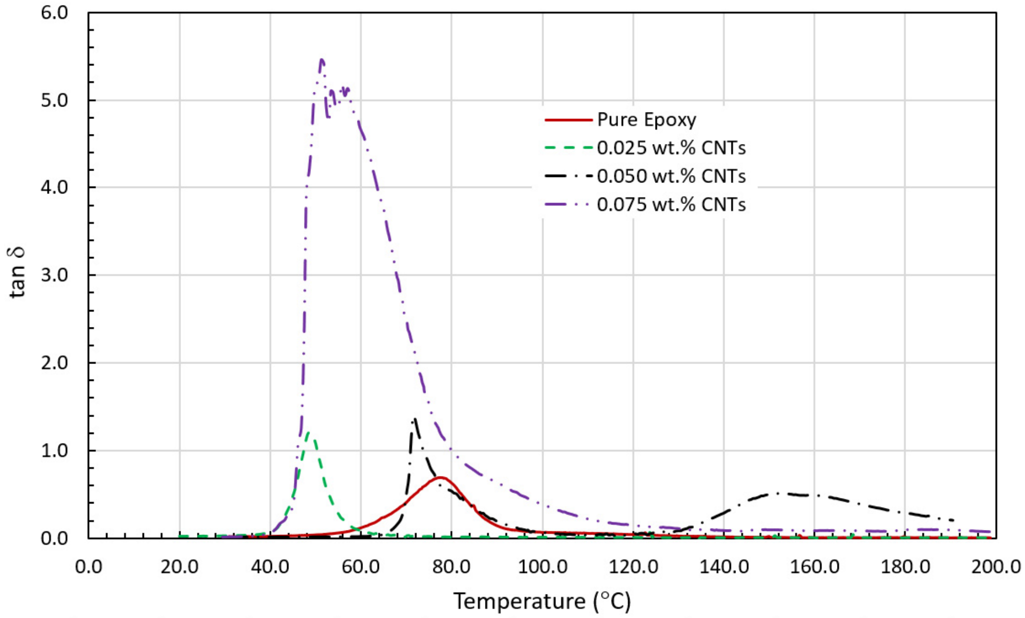

Figure 8. The effect of CNTs on tanδ of pure epoxy resin and nano-composite samples.

According to Figure 9, the maximum tanδ of the nano-composite sample reinforced with 0.075 wt.% CNTs increased by 696% compared to pure epoxy resin. Furthermore, the loss moduli of the samples reinforced with 0.025% and 0.05 wt.% CNTs increase by 74.9% and 99.5%, respectively, compared to the pure epoxy samples. An increase in tanδ indicates a higher ability of the material to dissipate energy. Since CNTs have the properties of energy absorption and folding due to the π-bond in their structure, by adding CNTs to the polymer matrix, the ability of the material to absorb energy increases, so tanδ increases [36].

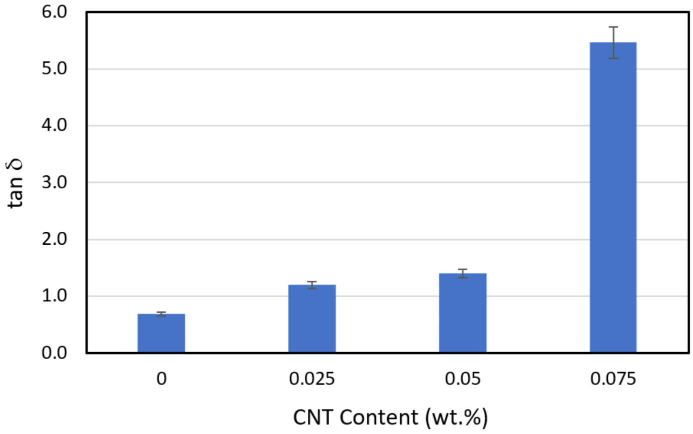

Figure 9. Comparison of tanδ of samples with different wt.% CNTs.

The above results are consistent with the research of Her and Lin [32]. They studied the mechanical dynamic properties by adding SWNTs to the epoxy resin and observed that by adding 1 wt.% CNT reinforcement to the epoxy, the storage modulus increased by 52%, and the loss modulus increased by 45% compared to the pure epoxy resin. Furthermore, research shows that adding 0.1 wt.% CNTs, the tanδ and the loss modulus of its epoxy samples were increased by 42% and 36%, respectively, compared to the pure epoxy resin [13].

3.2. Thermal Gravimetric Analysis (TGA)

TGA analysis was used to evaluate the effect of adding CNTs on the thermal stability of epoxy resin. Figure 10 shows the TGA plots of the pure epoxy resin and its nanocomposites reinforced with different wt.% CNTs. Moreover, Figure 11 shows a type of thermal analysis in which the rate of material weight changes upon heating is plotted against temperature (DTGA) for the same samples. From the figures above, the degradation temperature of the pure sample and the nanocomposite samples containing 0.025, 0.05, and 0.075 wt.% CNTs are different so that the degradation of the nanocomposite samples occurs at a higher temperature. In other words, by adding CNTs, we can conclude that the thermal stability of the nanocomposite samples is increased. The reason for this can be attributed to the very good thermal stability of CNTs. CNTs are excellent thermal conductors that allow applied heat to transfer through themselves so that the heat is not concentrated in one place, delaying degradation [37].

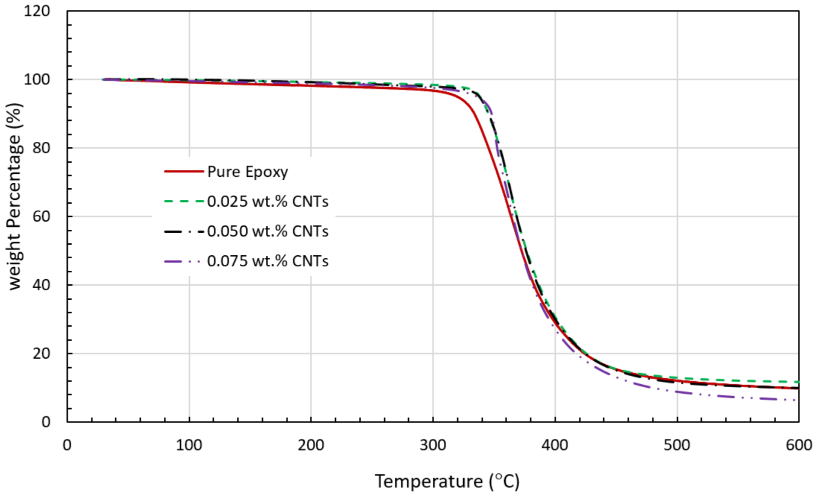

Figure 10. Comparison of TGA results of pure epoxy and nanocomposites containing different wt.% CNTs.

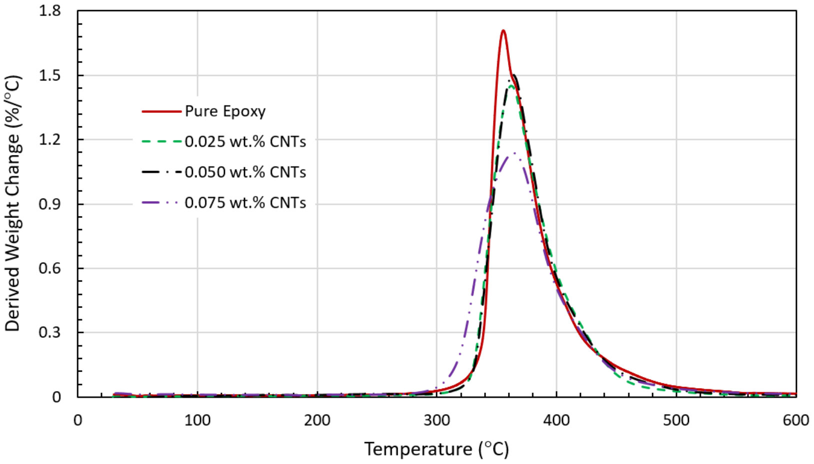

Figure 11. DTGA diagram of pure epoxy and nanocomposites containing different amounts of weight percentage of carbon nanotubes.

Table 2 shows the maximum weight loss temperature T_{max} values, which is defined as the maximum temperature of epoxy decomposition. With the addition of CNTs, the maximum weight loss temperatures increase due to the strong interactions between the CNTs and the epoxy matrix. At the same time, the CNTs act as physical crosslinks and reduce the fluidity of the polymer matrix; hence, the glass transition temperature increases [38].

Table 2. The maximum weight loss temperatures of nanocomposite samples, corresponding to the pure epoxy sample, containing different wt.% CNTs.

Sample	T_{max} (°C)
Pure Epoxy	355
Epoxy + 0.025% CNT	362
Epoxy + 0.05% CNT	363
Epoxy + 0.075% CNT	364

As can be seen from Table 3, the weight loss temperature of the nanocomposite samples containing 0.025, 0.05, and 0.075 wt.% CNTs equivalent to 10% weight loss of the epoxy sample increased by 11.4, 12.6, and 14.0 °C, respectively, compared to the pure epoxy sample. In addition, the epoxy samples containing 0.025, 0.05, and 0.075 wt.% CNTs corresponded to 50% weight loss of the epoxy sample, and the weight loss temperatures increased by 0.20, 3.8, and 4.3 °C, respectively, compared to the pure epoxy sample. The reason for this can be attributed to the very good thermal stability of CNTs and the surface interactions between CNTs and epoxy [39].

Table 3. Degradation temperatures of nanocomposite samples, corresponding to 10% and 50% weight loss for epoxy samples containing different wt.% CNTs.

Sample	T_{10} (°C)	T_{50} (°C)
Pure Epoxy	8.333	372.3
Epoxy + 0.025% CNT	2.345	372.5
Epoxy + 0.05% CNT	4.346	376.1
Epoxy + 0.075% CNT	8.347	376.6

A study on pure epoxy resin and CNT-reinforced epoxy matrix composites showed that by adding 1, 2, and 3 wt.% CNTs, the 5% mass loss temperatures of these materials were 351, 356, and 358 °C, respectively, which is comparable with this work on a similar model [40].

According to Figure 12, the thermal stability of the nano-composite samples reinforced with 0.025, 0.05, and 0.075 wt.% CNTs, compared to the pure epoxy sample at 10% weight loss of the sample, which increased by 3.4%, 3.7%, and 4.2%, respectively. In addition, compared to the pure epoxy sample at 50% weight loss of the epoxy sample, the thermal stability increasements of 0.04%, 1% and 1.1% were observed for the nanocomposite samples reinforced by 0.025, 0.05, and 0.075 wt.%, respectively.

Similar behavior to that reported by Loos [13] can be found in this work. His TGA analysis on reinforced epoxy composites with 0.1 wt.% and 0.25 wt.% CNTs showed that all samples began to lose weight around 70 °C, degraded by 90% around 550 °C, and the second weight loss occurred between 190–320 °C (peak at 250 °C), probably due to decomposition of lower molecular weight materials, and a third between 290–490 °C (peak at 370 °C).

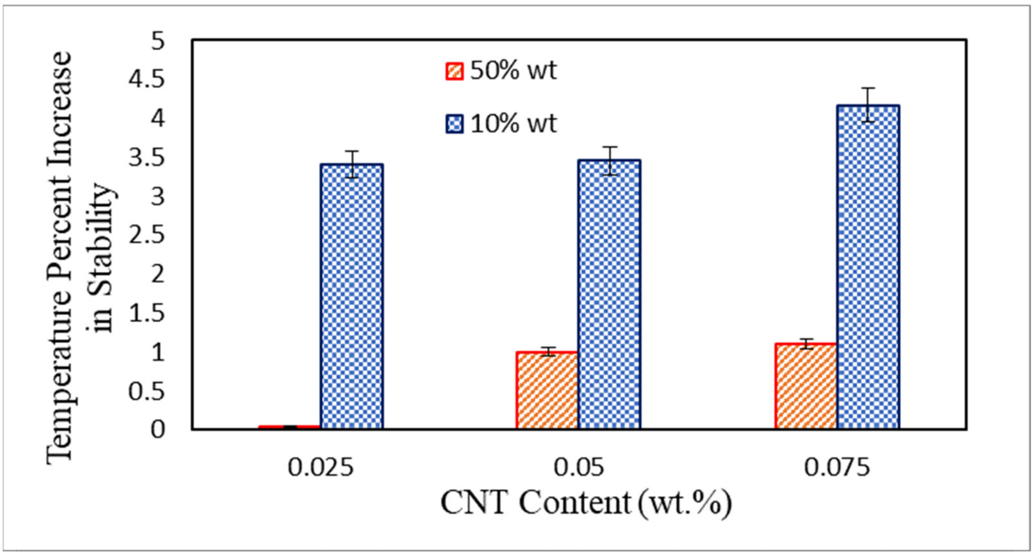

Figure 12. The percentage increase in thermal stability of the sandwich panels at 10% and 50% weight loss of the epoxy-containing different wt.% CNTs, compared to the pure epoxy sample.

3.3. Density

Density measurements were performed to evaluate the effect of adding CNT reinforcement on the density of the epoxy resin. It was observed that the density increased with the wt.% CNTs. According to Figure 13, the actual density of samples reinforced with 0.075 wt.% CNTs increased by 0.43% compared to the pure epoxy resin. Furthermore, the density of the samples reinforced with 0.025 and 0.05 wt.% CNTs increased by 0.172% and 0.35%, respectively, compared to the pure epoxy sample.

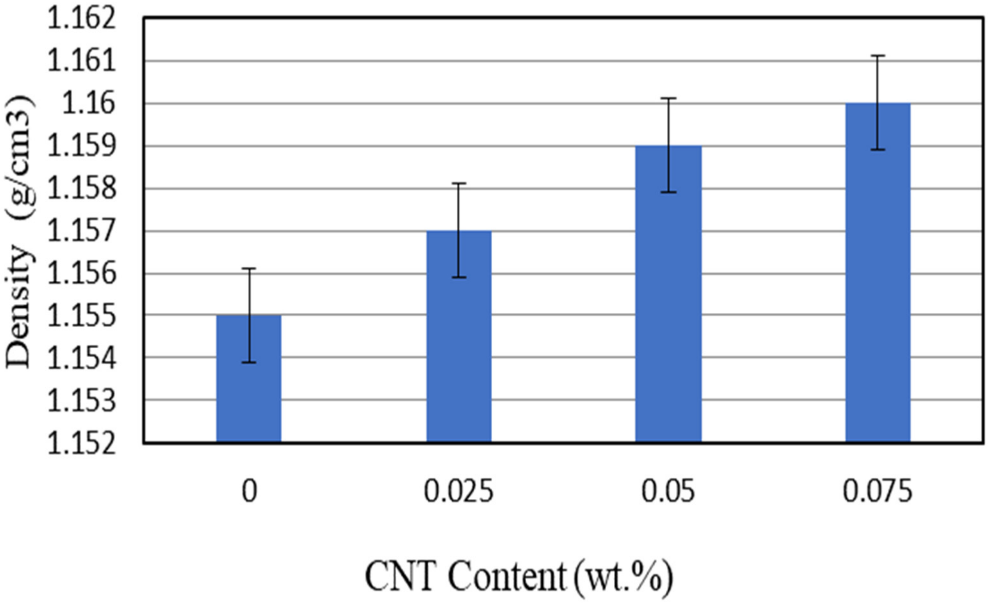

Figure 13. Effect of adding CNTs on the density of pure epoxy and nanocomposite samples reinforced with different wt.% CNTs.

In order to calculate the ratio of the actual density to the theoretical density of the materials, the theoretical densities of the CNT-reinforced epoxy matrix composites with different weight percentages of CNTs need to be calculated. Therefore, according to the law of mixtures as in Equation (6), the theoretical densities of the pure epoxy and nanocomposite samples reinforced with different wt.% CNTs, assuming that the theoretical density of the epoxy sample is 1.25 g/cm^3 and the theoretical density of CNTs is 1.6 g/cm^3, the theoretical densities of the nanocomposite samples reinforced by 0.025, 0.05, and 0.075 wt.% CNTs are calculated to be 1.2500, 1.2501, 1.2502, and 1.2503 g/cm^3, respectively [41].

$$\rho = \rho_e V_e + \rho_n V_n \tag{6}$$

where ρ_e and ρ_n denote the density of the epoxy and CNTs, respectively, and V_e and V_n are the weight percent of the epoxy and CNTs, respectively. The ratio of the actual density to the theoretical density for the pure epoxy and nanocomposite samples reinforced with 0.025, 0.05, and 0.075 wt.% CNTs are 0.9240, 0.9255, 0.9270, and 0.9278, respectively.

Ciecierska et al. [40] showed that the densities of the pure epoxy and CNT-reinforced epoxy matrix composites with 0.1, 1.0 and 2.0 wt.% of CNTs were 1.1972, 1.1994, 1.1996, and 1.1997 g/cm^3, respectively. The reason is that after the addition of CNTs in epoxy resin, the free movement of polymer chains is restricted because the free volume between macromolecules is occupied by CNTs; thus, the chains are more squeezed, and the crosslink density increases [40].

4. Conclusions

- DMTA showed that the storage modulus, loss modulus, and tanδ values increased with the increase of wt.% CNTs. The storage modulus, the loss modulus, and the tanδ of the nanocomposite sample reinforced with 0.075 wt.% CNTs increased by 43.8%, 74.4%, and 696%, respectively, compared to the pure epoxy resin. The reason why the three parameters of dissipation modulus, storage modulus and tanδ all increase with the increase of the wt.% CNTs is the energy absorption property of the CNTs.
- TGA showed that the epoxy nanocomposites were thermally stable due to the addition of CNTs. The increase in the degradation temperature of the nanocomposites is due to the strong interactions between the epoxy matrix and the CNTs, which is caused by the thermal stability of the CNTs. The pure epoxy sample has the lowest degradation temperature. By adding 0.075 wt.% CNTs to the epoxy resin, the temperature corresponding to a 10% and 50% weight loss in the pure epoxy samples increased by 14 °C and 4.3 °C, respectively. According to the results of the DTGA, the degradation temperature of the nanocomposite samples reinforced with 0.075 wt.% CNTs increased by 9 °C.
- Density measurements showed that the density of the nanocomposite samples increased by adding CNTs to the pure epoxy resin. The ratio of the actual density to the theoretical density of the nanocomposite samples reinforced with 0.025, 0.05, and 0.075 wt.% CNTs are 0.925, 0.926 and 0.927, respectively.
- The well-dispersed CNTs in the epoxy resin can strongly affect the performance of the composites. Otherwise, the CNTs cannot significantly improve the performance of the resin.
- Since CNT-reinforced epoxy matrix composites can absorb more energy through heat and can be used to reduce heat loss and resist high temperatures, which is an optimal choice for designers and engineers who want to increase the effectiveness of their products.

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