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Understanding Glass Transition Dynamics in Epoxy Reinforced with Hybrid Carbon Black and Silica Nanoparticles

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The development of nanocomposites with enhanced multifunctional properties is a critical focus in material science. This study addresses the inherent limitations of epoxy resins, such as thermal constraints, by incorporating hybrid carbon black (CB) and silica (SiO2) nanofillers. The aim was to investigate the effects of these hybrid reinforcements on the glass transition dynamics of epoxy nanocomposites. Utilizing a systematic approach, epoxy nanocomposites were prepared with varying concentrations and ratios of CB and SiO2. The glass transition dynamics were analyzed through differential scanning calorimetry (DSC) and Thermogravimetric analysis (TGA). Results demonstrated that the addition of 3 wt% SiO2 elevated the glass transition temperature (Tg) by 18°C. The hybrid combination of 4 wt% CB and 2 wt% SiO2 provided an optimal balance, achieving a significant improvement in the thermal properties. The study confirms that hybrid nanofillers can effectively enhance the multifunctional properties of epoxy nanocomposites. It is recommended that upcoming study be focus on application of these production process, researching the long-term durability of these nanocomposites, and utilizing other potential hybrid systems to supplement optimum performance.

Keywords: Epoxy, Nanocomposite, Carbon Black, Silica.

1. Introduction

The continuous improvement of material science has motivated the growth of innovative nanocomposites with improved multifunctional properties, mostly in electrical conductivity and thermal stability. Epoxy resins is related to the class of exceedingly cross-linked thermoset polymers utilized most frequently with reinforcing fibers in an extensive variety of composite applications [1-3], The high crosslink density makes them inherently brittle materials leading to deficient fracture energy [1]. Epoxy resins, known for their excellent mechanical properties, chemical resistance, and strong adhesion, are widely used in various industrial applications such as coatings, adhesives, and composite matrices [4-6]. The dispersion of nanoparticles in the material matrix is primarily used to increase or advance the properties of the composite material such as electrical properties, mechanical strength, wear resistance, toughness, thermal properties, Optical properties, and others [7]. А composite can be classified as a nanocomposite if at least one of the reinforcement

dimensions is in the nanometer range (1–100 nm) [8, 9], The addition of rigid fillers like carbon black (CB) [7, 10-13], cobalt ferrites nanofiller [14], multiwalled carbon nanotubes (MWCNT's) [10, 15], titanium (TiO₂) [16], Alumina (Al₂O₃) nanoparticles [17, 18], Glass [17], and silica (SiO₂) nanoparticles [15, 17, 19, 20] can improve the strength and modulus of epoxy nanocomposites while also increasing the fracture toughness without decreasing the glass transition temperature of the nanocomposites. Recently, hybrid nanofiller loading has been utilized to fabricate multifunctional composites.[14] and plays a vital role in everyday life [21].

The Glass Transition Temperature (Tg), should not be confused with melting point (Tm), It is the temperature range where a thermosetting polymer changes from a hard, rigid or "glassy" state to a more flexible, yielding or "rubbery" state. The Glass Transition Temperature (Tg) is one of the most important properties of any epoxy and is the temperature range over which the mobility of the

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polymer chains increases significantly [22]. The ultimate (Tg) is determined by several factors: the chemical structure of the epoxy resin, the type of hardener and the degree of cure and the resulting epoxy must not be subjected to condition above it's Tg. Kozako et al. [18] reported a constant glass transition temperature when they dispersed varied percentages (3-10 %wt) of Al₂O₃ in the epoxy matrix (Epikote 816B) with Epicure 11 as a hardener, In contrast, Liang, M. and K.L. Wong [23] their study improved the Tg of epoxy with 1 %wt of Al_2O_3 . Similarly Lavorgna et al. [24] improved the glass transition temperature of epoxy by about 20 °C with silica-enriched carbon nanotubes they attributed this increase to fair even dispersion of the nanofiller due to surface functionalization of the nanofiller which reduces agglomeration of the nanofiller owing to the high surface area [25], it is therefore established that the glass transition temperature can be improved by carefully dispersing the appropriate percentage weight of specific nanoparticles [26-28] and depends on the effect of the filler size, filler loading, and dispersion conditions of the nanofillers [29]. Among the numerous nanofillers investigated, carbon black and silica nanoparticles have garnered considerable attention due to their distinct and complementary properties. Carbon black, with its high surface area and conductive nature, significantly enhances the electrical conductivity of polymer composites. On the other hand, silica nanoparticles are known for their ability to improve the thermal stability and mechanical properties of the host matrix. Previous studies have primarily focused on the individual effects of carbon black or silica nanoparticles on the properties of epoxy nanocomposites. However, the synergistic effects of hybrid nanofillers, combining both carbon black and silica remain largely unexplored. The potential to tailor the thermal properties through hybrid reinforcement offers a to novel approach developing advanced nanocomposites with superior performance. we emphasized the exploration of synergistic effects between carbon black (CB) and silica (SiO_2) nanofillers, which are largely unexplored in epoxy systems.

This study aims to investigate the effects of hybrid carbon black and silica nanofiller reinforcement on the electrical conductivity and glass transition dynamics of epoxy nanocomposites. By systematically varying the concentration and ratio of these nanofillers, we seek unravel the interactions between carbon black and silica within the epoxy matrix and their collective impact on the material properties. This is carried out by employing a response surface methodology (RSM) with a Box-Behnken design, we seek to understand the individual and interactive effects of key processing parameters such as mixing speed, mixing time, carbon black weight fraction, silica weight fraction, and the state of carbon black nanoparticles (as received, thermally treated, and thermally and chemically treated) on the glass transition temperature and the electrical conductivity of the epoxy nanocomposite. We elaborated on the significance of using response surface methodology (RSM) with a Box-Behnken design. This approach allowed for the systematic optimization of key processing parameters (such as mixing speed, mixing time, and filler weight fractions), enhancing our understanding of the interactive effects of CB and SiO₂ on thermal properties. To achieve this, a methodology comprehensive was emploved. involving the preparation of epoxy nanocomposites with varying concentrations and ratios of carbon black and silica nanoparticles. The glass transition dynamics were analyzed through differential scanning calorimetry (DSC) and Thermogravimetric analysis (TGA), The glass transition temperature naturally manifests itself as a change of slope in a DSC thermogram. A simple way to determine the glass transition temperature is to draw a tangent on the initial straight-line representing baseline and draw another tangent on the slope. The crossover point is the glass transition temperature. These techniques provide a detailed understanding of the thermal and mechanical behavior of the nanocomposites. In this research, we present a comprehensive analysis of the thermal properties of the hybrid nanocomposites. The methodology, results, and implications of these findings are detailed, culminating in recommendations for future research directions in the field of hybrid nanocomposite materials.

Methodology Materials

The epoxy resin used was supplied by Smooth-On with the commercial name of EpoxAmite 100 Resin and 102 medium hardeners. The 102 medium hardener consists of triethylenetetramine (TETA) with propylene oxide (50-55 %), polyoxopropylene triamine (POPTA) (20-25 %) and TETA (15-20 %). The CB nanoparticle was obtained from CABOT under the commercial name of VULCAN[®] XC72 with a volume resistivity of 6 Ω -cm at 23 °C. VULCAN[®] XC72 has an iodine number of 253 mg/g density of 1.7-1.9 g/cm³ and OAN of 174 cc/100g,

The SiO₂ nanoparticle with 10-25 nm diameter was obtained from Zhengzhou Dongshen Petro Chemical Technology Co, Ltd. The analytical grade chemicals

used for surface treatments of the nanoparticles include tetrasodium pyrophosphate (TSPP), formic acid (FA), 3 Isocyanatopropyltriethoxysilane (IPTES), 3-Aminopropyltrimethoxysilane (APTES) and isophorone diisocyanate (IPDI).

2.2 Production of Epoxy Resin/ Carbon Black Nanocomposite

Three variations of CB which include as received CB, thermally treated CB and chemically modified thermally treated CB are used as one of hybrid nanofillers. 170g of epoxy was poured into beaker and preheated for 30 minutes at 40 °C to improve viscosity.

20% nano silica with respect to CB %wt was measured and added to the beaker containing 170g of epoxy. 10 ml of IPDI was measured using a syringe and placed into the epoxy containing beaker, also added was 1.5 ml of IPTES. The mixture was stirred manually until a homogenous mixture was achieved. 12.5 %wt of CB with respect to epoxy resin content was placed into another beaker containing 250 ml of acetone, 4.6 % of APTES with respect to %wt of CB was also added. 5 %wt of TSPP with respect to CB %wt was added to the acetone containing beaker, FA was added gradually using a syringe until the PH value of the mixture is 4. The content of the beaker is manually mixed before the sonication process. Sonication was done with an ultrasonic cleaner model 008 with an ultrasonic power of 50 w and a frequency of 40 KHz. The sonication process proceeded for 15 minutes at a time and a break period of 2 minutes between intervals. After 60 minutes of sonication, the content of the beaker containing acetone was mixed with the content of epoxy-containing beaker in a new beaker, the content is stirred manually until a homogenous mixture was achieved.

The beaker was then placed on a magnetic stirrer, the temperature was set to 60 °C and the speed was adjusted to 950 rpm. The magnetic stirring was made for 60 minutes while maintaining the temperature at 60 °C to ensure proper dispersion. Further stirring was done by the use a mechanical mixer at 600 rpm for 30 minutes; the mechanical mixer use was a pensonic PM-112 hand mixer with an electrical power of 120 w and a 5-speed selection. After the mixing process, acetone was evaporated using a hot air blower and by increasing the temperature of the magnetic stirrer to 75 °C to guarantee thorough evaporation of the solvent. The sample was then placed in the degassing chamber; the vacuum pump was operated for 30 minutes to remove air bubbles trapped within the sample. The resin-hardener ratio of 100:29 as advised by the manufacturer was adopted; 49.3g of hardener was added to the degassed sample and blended manually till a homogenous mixture was attained. The nanocomposite was then transferred into the molds. The samples were post-cured for 60 minutes at 100 °C in an oven to complete the curing action. processing parameter of the epoxy The nanocomposite is shown in Table 1

 Table 1: Processing Parameter for the production of Epoxy Nanocomposite

| Samples | CB %wt | CB600 %wt | CB-M %wt | SiO₂ %wt | Speed Rpm | Hours |
|---------|--------|--------------|-------------|-------------|--------------|-------|
| EP | - | - | - | - | - | |
| EP2 | 12.5 | - | - | - | 700 | 2.50 |
| EP9 | 5 | - | - | 10 | 700 | 1.00 |
| EP12 | 12.5 | - | - | 10 | 950 | 2.50 |
| EP32 | - | - | 12.5 | 20 | 1200 | 2.50 |
| EP46 | - | 5 | - | - | 950 | 2.50 |
| EP50 | 5 | - | - | - | 950 | 2.50 |
| EP57 | 12.5 | - | - | - | 950 | 1.00 |
| EP71 | 12.5 | - | - | - | 950 | 4.00 |
| EP75 | 12.5 | - | - | - | 1200 | 2.50 |
| EP82 | 12.5 | - | - | - | 700 | 2.50 |

2.3 Surface Treatment of Nanofillers

The surface treatment of carbon black (CB) and silica (SiO_2) nanofillers was conducted to enhance their dispersion in the epoxy matrix and improve interfacial bonding. Specific chemical additives were used during the treatment process to achieve functionalization and reduce agglomeration:

2.3.1. Isocyanatopropyltriethoxysilane (IPTES):

IPTES was used as a coupling agent to introduce functional silane groups onto the surface of the silica nanoparticles. These groups improve compatibility between the inorganic SiO_2 and the organic epoxy matrix by forming covalent bonds at the interface. This enhances interfacial adhesion and ensures better stress transfer within the composite. Additionally, IPTES contributes to forming a stable dispersion preventing nanoparticle by agglomeration.

2.3.2. Formic Acid (FA):

FA was employed to adjust the solution's pH during the surface treatment process, optimizing the reaction environment for functionalization. By maintaining a slightly acidic pH, FA facilitates the removal of surface impurities and activates the nanoparticles, making them more receptive to chemical bonding. This results in cleaner, more reactive surfaces, further enhancing their compatibility with the epoxy matrix.

2.3.3. Tetrasodium Pyrophosphate (TSPP):

TSPP acts as a dispersant to stabilize the nanoparticles in solution. It prevents the aggregation of nanofillers during the mixing process, ensuring a more uniform distribution throughout the epoxy matrix.

These surface treatments collectively aim to improve the dispersion of the nanofillers, reduce interfacial tension, and enhance the overall performance of the hybrid epoxy nanocomposite. The treated nanofillers demonstrated improved mechanical, thermal, and electrical properties compared to untreated counterparts.

2.4 Experimental Design

Response surface methodology was adopted to optimize the production of the nanocomposite; the empirical model was obtained using Box Behnken experimental design. The experimental factors and their levels are as shown in Table 2

Table 2: Experimental Factors and their Levels

| Levels of Variables | Input Variables | | | | | | | |
|------------------------|------------------------------------|----------------------------------|-----------------------------------|---------------------------------------|--|--|--|--|
| | Magnetic Stir Speed (rpm) | Magnetic Stir Time (Hours) | CB Weight Fraction (%Wt) | Silica Weight Fraction (%wt) | State of CB Nanoparticle | | | |
| -1 | 700 | 1 | 5 | 0 | As Received [1 0] | | | |
| 0 | 950 | 2.5 | 12.5 | 10 | Thermally Treated [01] | | | |
| +1 | 1200 | 4 | 20 | 20 | Thermally & Chemically treated [-1 -1] | | | |

One (1) categorical and four (4) numeric experimental factors (state of CB nanoparticle, magnetic stir Speed, magnetic stir time, CB weight fraction and silica weight fraction) were used in the research as shown in Table 2. The response was measured according to the standards specified in Table 3. A total of 87 runs in a single block were used with 5 central points for each categoric factor as is typical with Box-Behnken experimental design. Some of objectives of this research are to determine which factors are most significant on the response variables, and to determine where to set the dominant controllable factor so that the response is almost near the desired optimal value. The Design-Expert 10 software was used in the generation of the experimental design and the analysis of the interactions of the factors

Table 3: Response Variables

| S\No. | Response | Unit | Standard |
|-------|-------------|------|----------|
| 1 | Glass | °C | |
| | Transition | | |
| | Temperature | | |

3. Results and Discussion

3.1. Characterization of the Nanoparticles and Nanocomposites

The properties of the nanocomposite are greatly influenced by the loading and degree of dispersion of the nanoparticles. The interfacial interaction between the matrix and the nanofiller is affected by the size, morphology and surface chemistry of the dispersed nanoparticle. Due to its high thermal conductivity, CB can help distribute and dissipate heat more evenly across the composite. By conducting heat away from localized hotspots, CB reduces the likelihood of thermal breakdown in specific sites within the matrix, thereby enhancing the composite's resistance to thermal stresses. When CB forms a percolation network within the epoxy matrix, it provides a structurally reinforcing framework that improves the material's rigidity and helps resist thermal degradation. This network restricts the mobility of polymer chains, thus limiting their ability to break down under heat. When CB is used alongside other fillers like silica, CB can further improve the thermal stability of the composite. The silica particles stabilize the matrix by reducing polymer chain mobility, and CB's network structure adds additional heat-dissipative properties, creating a synergistic effect that enhances the overall thermal performance.

Silica nanoparticles, in contrast, are highly stable, rigid, and possess a high surface energy that improves thermal and mechanical properties. Their ability to form strong interfacial bonds with the epoxy matrix aids in stress transfer and prevents agglomeration of CB particles, which can otherwise compromise the material's overall stability. The high thermal stability of SiO₂ also helps to elevate the glass transition temperature (Tg), as it restricts molecular motion at elevated temperatures. The addition of SiO₂ thus contributes to a more thermally stable composite with a reinforced network that can withstand greater thermal stresses without losing structural integrity.

The synergistic interaction between CB and SiO₂ is particularly beneficial in terms of Tg improvement. Silica particles create a "nano-confinement" effect within the epoxy matrix, limiting the free volume and restricting the mobility of polymer chains, which contributes to a higher Tg. Simultaneously, the welldispersed CB network, aided by the silica's ability to prevent particle agglomeration, promotes a uniform distribution of thermal and mechanical stresses throughout the matrix. This balance of rigidity from SiO₂ and conductivity from CB produces a composite material with improved thermal stability and electrical conductivity, making the hybrid approach superior to single-filler systems.

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The influence of thermal and chemical treatment on the properties of the as received Carbon Black (CB) nanoparticles can be explained from the XPS results of the nanoparticle and the nanocomposites. The properties of the thermally treated carbon black (CB600), the silane or chemically treated carbon black (CB-M) nanoparticle is presented.

3.1.1. XPS Analysis

The XPS characterization was done for a spot size of 400 μ m and for step energy of 1.00 eV for a total energy steps of 1361 for both the nanoparticles and the formed nanocomposite.

3.1.1.1. XPS of the Nanoparticles

The surface chemistry of as received CB nanoparticle at a depth of 10 nm and a spot size of 400 μ m was conducted, the survey scan indicates the presence of dominant Carbon (C1s) element at a binding energy of 284.48 eV.

The CB nanoparticle was treated to improve its dispersion and interaction with epoxy resin, the effect of the thermal and chemical modification can be seen as shown in Table 4. There is a noticeable reduction in the percentage composition of fluorine with respect to increase in the temperature of the thermal treatment; this is as a result of the oxidation effect of increase in temperature during thermal treatment. Table 4 shows the oxidation effect of the nanoparticle with the maximum increase of oxygen at 61.86 % when CB600 was chemically modified from 0.9 -2.36 of C1s. There is an increase in the number of oxygen available to bond with hydrogen thereby improving the hydrophilicity of the CB nanoparticle; hydrophilicity does improve the dispersion and wettability of the nanoparticle in epoxy resin. consequently, there is an increase in the area of the O1s curve as the treatment progress. The curve also indicates other functional groups as shown in Table 4 showing the existence of additional Si2p and N1s from the alkosilane and the amino groups of APTES.

Table 4: Surface Elemental Composition of the CBnanoparticles

| Samples (%) | C1s | F1s | O1s | Na1s | N1s | S2p | Si2p |
|-------------|-------|------|------|------|------|------|------|
| СВ | 97.59 | 1.20 | 0.90 | _ | — | 0.31 | — |
| CB350 | 98.22 | 0.50 | 0.90 | - | — | 0.38 | - |
| CB450 | 98.19 | 0.57 | 1.01 | _ | _ | 0.23 | _ |
| CB600 | 97.89 | 0.71 | 1.06 | - | — | 0.34 | - |
| CB-M | 95.68 | | 2.36 | 0.44 | 0.72 | 0.25 | 0.54 |

3.1.1.2. XPS of the pure epoxy and the Epoxy nanocomposites

The XPS of the pure and cured epoxy (EP) sample shows a carbon (C1s) content of 54.34 % at a binding energy of 294.08 as shown in Table 5, this percentage differ from the epoxy-carbon (EP-CB) black nanocomposite with 59.68 % which is due to the presence of CB-M. The addition of SiO2 results in well dispersed C1s with consequent increase in C1s content to 61.24 % over the 400 µm spot.

| Table | 5: | Surface | Elemental | Composition | of |
|--------|------|---------|-----------|-------------|----|
| Nanoco | mpos | ite | | | |

| Samples (%) | C1s | O1s | N1s | Cu2p3 | Si2p |
|-------------|-------|-------|------|-------|------|
| EP | 54.34 | 33.38 | 4.49 | 2.54 | 4.82 |
| EP-CB | 59.68 | 31.08 | 5.37 | — | 3.87 |
| EP-CB-Si | 61.24 | 30.12 | 6.69 | — | 1.94 |

3.3. Statistical Analysis of the Glass Transition Temperature (Tg) Data

The Box-Cox transformation report from the software as shown in Figure 1 recommended a natural log transformation. The transformation is used to modify the distributional shape of the glass transition temperature data to be more normally distributed so that tests and confidence limits that require normality can be appropriately used. The current lamda position is at 1 as shown in Figure 1 which is outside the low confidence interval of -0.58 and the high confidence interval 0.80.

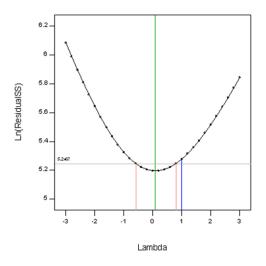


Figure 1: Box-Cox Plot for Power Transformation Applied to the Glass Transition Temperature Response Model

| Source | Std. | R-Squared | Adjusted | Predicted | PRESS | 6 |
|-----------|--------------|---------------|---------------|------------------|-------------|-----------|
| | Dev. | | R-Squared | R-Squared | | |
| Linear | <u>0.037</u> | <u>0.9492</u> | <u>0.9454</u> | <u>0.9399</u> | <u>0.13</u> | Suggested |
| 2FI | 0.035 | 0.9620 | 0.9504 | 0.9340 | 0.14 | |
| Quadratic | 0.034 | 0.9670 | 0.9542 | 0.9361 | 0.14 | |
| Cubic | 0.026 | 0.9896 | 0.9736 | 0.9586 | 0.089 | Aliased |

Table 6: Summary Statistics of Impact Strength Model

From Table 6, PRESS for the linear model is 0.13; the model has an R-square of 0.9492, the predicted R-square is 0.9399 and is in logical agreement with the adjusted R-square since their difference is less than 0.2. The linear model equations for the conductivity are

Ln(Y_tg (CB))=+3.13+9.56E^(-003)*A+6.027E^(-005)*B+0.03*C+0.01*D-1.53E^(-004)*CD (1)

Ln(Y_tg (CB600)=+3.24+9.56E^(-003)*A+6.027E^(-005)*B+0.03*C+0.01*D-1.53E^(-004)*CD (2)

Ln(Y_tg (CBM)=+3.36+9.56E^(-003)*A+6.027E^(-005)*B+0.03*C+0.01*D-1.53E^(-004)*CD (3)

Where

Ytg(CB) : Glass Transition Temperature of Epoxy-CB Nanocomposite sample

Ytg(CB600) :Glass Transition Temperature of Epoxy-CB600 Nanocomposite sample

Ytg(CB-M) : Glass Transition Temperature of Epoxy-CB-M Nanocomposite sample

A, B, C, and D are as previously defined

Table 7: ANOVA Table for the Regression Model of Equation (1) - (3)

| Source | Sum of | | Mean | F | p-value |
|--------------------------|------------|----|------------|--------|----------|
| | Squares | Df | Square | Value | Prob > F |
| Model | 2.04 | 9 | 0.23 | 172.60 | < 0.0001 |
| A-Magnetic. Stir Time | 7.404E-003 | 1 | 7.404E-003 | 5.65 | 0.0119 |
| B-Magnetic Stir speed | 8.154E-003 | 1 | 8.154E-003 | 6.22 | 0.0148 |
| C-CB content | 1.30 | 1 | 1,30 | 993.79 | < 0.0001 |
| D-Si Content | 0.25 | 1 | 0.25 | 191.17 | <0.0001 |
| E-Modification | 0.46 | 2 | 0.23 | 175.37 | <0.0001 |
| CD | 1.588E-003 | 1 | 1.588E-003 | 1.21 | 0.2744 |
| CE | 6.061E-003 | 2 | 3.031E-003 | 2.31 | 0.1059 |
| Residual | 0.10 | 77 | 1.311E-003 | | |
| Lack of Fit | 0.084 | 65 | 1.299E-003 | 0.95 | 0.5884 |
| Pure Error | 0.016 | 11 | 1.371E-003 | | |
| Cor Total | 2.14 | 86 | | | |

ANOVA was used to check the adequacy of the model for the responses in the experimentation. The F-value of the quadratic model is 172.60 as shown in Table 7. There is only a 0.01% chance that an Fvalue this large could occur due to noise. From Table 6 terms A, B, C, D and E were significant since their p-value (Prob> F), are very small pvalues (p< 0.05). The R-square of 0.9492 showed that the unpredictability in the glass transition temperature of 94.92 % could be explained by the quadratic equations and only 4.18 % of the total glass transition temperature variations were not explained by the independent variables of the quadratic equations. Figure 2 shows the measured values versus the predicted values of conductivity of the nanocomposite from the linear model of equations 1 – 3. The plot shows uniformly scattered points along the regression line which is an indication of a good prediction.

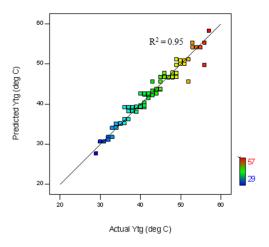


Figure 2: Plot Comparing Predicted and Measured Glass Transition Temperature (Tg) Values

Changes in temperature can have an enormous impact on the performance properties of epoxy and glass transition temperature (Tg) is one of the most important properties of any epoxy and is the temperature region where the polymer transitions from a hard, glassy material to a soft, rubbery material. It is therefore very important to ensure that the operating temperature of the epoxy does not exceed it's Tg.

Figure 3 shows the effect of interaction between SiO2 content and CB content on the glass transition temperature of the nanocomposite while keeping the other parameters at their mean position. The Tg is observe to increase with CB and SiO2 %wt loading, the interaction between the CB and SiO2 %wt increased the Tg to 59 0C. The increase in Tg due to nanoparticle loading can be explained by the interfacial constrains within the nanocomposite.

When nanoparticles are dispersed in epoxy matrix, a small, extremely immobilized layer of a few nanometer-thick is developed near the surface of the nanoparticles. The formation entropy and the chains kinetics of this interaction region of the epoxy layer surrounding the nanoparticles are severely altered [30]. As the filler content increases the volume fraction of the interaction region in the nanocomposites increases. The epoxy chains in this region are under constrain because of the interfacial epoxy-nanoparticle interactions thereby increasing the Tg of the nanocomposites.

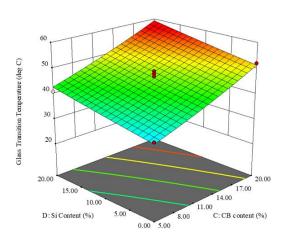


Figure 3: Response Surface Plot of Glass Transition Temperature (Tg) for Epoxy/CB-M Nanocomposite at 950 rpm Stir Speed and 2.50 Hours Stir Time

Figure 4 shows the interaction effect of CB-M content and modification of CB on the glass transition temperature of the nanocomposite. An increase in the Tg of the nanocomposite is observed as the treatment of nanoparticle progress from thermal treatment to thermal-chemical treatment. The high values for the Tg may be related to improved interfacial interaction due the of CB-M functionalization nanoparticles. the hardener molecules covalently linked to an epoxy system increase the mobility of the polymer chains, favouring the thermal transfer between the CB-M and the resin, which leads to a high Tg.

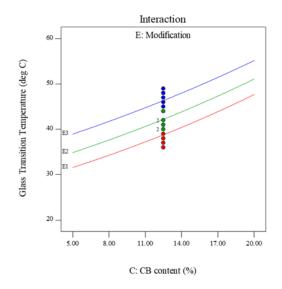


Figure 4: Interaction Effect of CB-M Content and Carbon Black Modification on Glass Transition Temperature (Tg) of Epoxy Nanocomposites

3.4. Long-Term Stability of the Nanocomposite

While a comprehensive evaluation of long-term stability was beyond the scope of this study, several factors suggest that the hybrid carbon black (CB) and silica (SiO_2) nanocomposites are likely to exhibit enhanced thermal and structural stability over extended periods. The combination of CB and SiO₂ contributes to a robust and synergistic reinforcement network within the epoxy matrix, which has been shown to delay thermal degradation in similar systems[10].

The addition of SiO₂ nanoparticles is particularly glass increasing the effective in transition temperature (Tg), which reduces the likelihood of polymer softening or thermal breakdown during prolonged exposure to elevated temperatures. Furthermore. the chemically treated CB nanoparticles improve electrical conductivity and enhance the nanocomposite's mechanical and thermal stability by forming stable interfacial bonds with the epoxy matrix. These interfacial bonds are less prone to degradation, as evidenced by the material's improved Tg and reduced thermal expansion coefficients observed in this study.

Studies on similar hybrid systems [5] have reported that including inorganic nanofillers, such as silica, significantly mitigates matrix degradation by limiting chain mobility and acting as a thermal barrier. Additionally, the uniform dispersion of nanofillers achieved through surface treatment processes reduces the risk of microcracking and delamination, further contributing to long-term performance. Future work can include an experimental analysis of the nanocomposite's thermal stability under prolonged thermal and mechanical cycling conditions. This will provide more definitive insights into the material's durability for high-performance applications, such as aerospace, automotive, and electronics.

5. Conclusion

In this study, the hybrid reinforcement of carbon and silica nanoparticles black in epoxy nanocomposites was investigated, revealing significant enhancements in thermal stability. Carbon black and silica nanoparticles elevated the glass transition temperature (Tg), thus improving thermal stability. The synergistic combination of these nanofillers resulted in superior properties compared to individual fillers, highlighting the importance of proper dispersion and interaction within the matrix. Utilizing response surface methodology with a Box-Behnken design the research effectively optimized the concentrations ratios of the nanofillers, and providing а comprehensive understanding of their interactions. These findings demonstrate the potential of hybrid nanofillers in creating advanced epoxy nanocomposites with thermal stability I properties suitable for various industrial applications. Future research should focus on scaling up production, investigating long-term durability, and exploring other hybrid systems to further enhance these properties.

Authors' Contributions

The idea was conceived by the sole author, conducted and written by the same author

Conflict of interest

The authors declare no conflict of interest.

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