Hybridization effect on Electrical Conductivity and Thermal Behavior of Epoxy/Carbon Nanotubes and Biosynthesized Silver Nanoparticles Composites

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Abstract

An attempt was made in this work to decorated carbon nanotubes (CNTs) in a polymer matrix using biosynthesized silver nanoparticles (GAgNPs) using Cashew leaves as a reduction agent. The new hybrid epoxy-CNTs+ GAgNPs composites were produced by modified solution-stir-cast method. The microstructure, thermal properties, strength, and electrical conductivity of the produced composites were determined. The electrical conductivity of the epoxy polymer has been enhanced from 5.6x10^-13S/cm to 4.80x10^-3S/cm for epoxy-0.5%CNTs and 9.1x10^-3S/cm epoxy-0.5%CNTs-0.5%GAgNPs. GAgNPs was effective used to improve the strength of conducting epoxy-CNTs for electronic devices. The addition of CNTs and GAgNPs to epoxy increases the glass transition temperature. It was established that GAgNPs can be promising materials to enhanced thermal conductivity, strength, electrical conductivity of epoxy-CNTs and recover the potential reduction for electronic devices application.

1.0 Introduction

The combination of high electrical conductivity and thermal properties are new hybrid composites used in the development of conductive adhesives of electronic devices such as multichip, printed circuit boards, and metallic packages [1,2]. A carbon nanotube (CNTs) is one of the nanoparticle's electrical conductive reinforcement that is attracting the attention of researchers due to the high electrical conductivity of CNTs[3,4]. The addition of CNTs to epoxy changes the electrical insulating properties of epoxy to conductive polymer[5]. Although with the high electrical conductivity and thermal of CNTs there is a dispersion of entanglement of the CNTs during the production and high cost of CNTs limited there used in electronic devices [6]. A great shear force is usually needed to help disperse CNTs and avoid entanglement in the polymer matrix. This limitation is still a challenge in the development of polymer/CNTs composites[7].

The modification of CNTS such as oxidation in acid solution [8-9], metal plating Surface[10], plasma treatments [11] dry oxidation in oxygen[13] and metallic nanoparticles [13] has been successfully used to enhanced the functionality of CNTs in polymer. Silver nanoparticles (AgNPs) [14], carbon black [15] and ionic liquid [16] has been successful added to CNTs to produced conductive composites. AgNPs have good thermal conductivities and electrical properties and they are used as catalysis, antimicrobials, electronic devices and conductive inks[17]. The used of AgNPs inhibits the formation of a nanostructured conductive and enhanced the electrical properties of polymer [18-19].

AgNPs +CNTs has find used in area of high electrical conductivity and thermal behavior nanoparticle: Ma et al [14] presented the enhancement of the electrical conductive of epoxy-carbon nanotube-coated silver nanoparticles. The microstructure, mechanical, thermal, and electrical properties of the developed composites were determined. The mechanical properties of epoxy-CNT and epoxy-Ag@CNT were closed, with enhanced electrical conductivity in the epoxy-Ag@CNT nanocomposites. Montazeri and Chitsazzadeh [20] reported on the thermal and mechanical properties of CNTs/epoxy composites. The
authors observed a change in elastic modulus from -14.6 to 46.5% for pure epoxy polymer with raises in Tg(6.6°C).

Dorigato et al.[15] improved the electrical conductivity of epoxy/ carbon fibers, carbon black in –situ in silver nanoparticles. They observed in the decrease in glass transition temperature after the addition of carbon fibers and carbon black. Ag nanoparticles added to epoxy/ carbon fibers+ carbon black help to stable temperature of the elevated voltage. From the literature, it was observed that the used of AgNPs to enhance the electrical conductivity of polymer/CNTs is promising, however the cost of silver is higher than the cost of CNTs, and this further restricts the application of polymer/CNT+AgNPs. The high cost of silver used in improving the electrical conductivity of epoxy/CNTs that motivated interest in the development of epoxy/CNTs +GAgNPs nanocomposites based on AgNPs biosynthesis from plant extract. Also the production of AgNPs using plant extracts and there used in the biological field has been documented in literature [21-24], there is little or no information of the potential application of GAgNPs in the composites industry, hence this work will report for the first time the thermal properties and electrical conductivity of epoxy/CNTs +GAgNPs nanocomposites.

Biosynthesis of nanotechnology has received great attention in recent, this was as a result of sustainable commercial viability, eco-friendly and safer for all beings with unique properties [25-27]. In this work, Cashew leaves extracts was considered as a redacting for the biosynthesis of GAgNPs, Researchers [29-30] have reported that Cashew leaves are acidic in nature and contains anacardic acids which are antibacterial agents that is used in the redaction of Ag⁺ to Ag. Cashew product has been previously exploited in nanotechnology [30]. The use of green plant extract to synthesize AgNPs may not only be less costly by also more eco-friendly.

2.0 Materials And Methods

The CNTs were purchased from Hongwu International Group of Company China. The CNTs are multi walled CNTs with average diameters of 10-40nm and 10-20µm length (see Figure 1).

The GAgNPs were synthesized using cashew leaves. The fresh cashew leaves used in this work were obtained from the surrounding of the University of Nigeria Nsukka Nigeria. The fresh leaves were washed with distilled water and dried. 100ml of ethanol was then added to the washed cashew leaves and left for 1hr, after which a solution of AgNO₃(100ml) was added. The addition of AgNO₃ to the cashew leaf extract samples produced an instant color change from an original, yellow solution to a dark brown solution (see Figure 2a). The reaction was left for 30min before heating in an electric oven at 100°C under stirring with a speed of 2000rpm, after heating the solution was centrifuge for 1hr to obtain the solid GAgNPs.

The functionalization of the CNTs was done using 65%HNO₃ and 30%H₂O₂. The CNTs were first treated with 100ml for 5hrs, and then deionized water was added to the solution and was filtrated. The filtered was diluted to obtain a Ph of neutral. The H₂O₂ was added to obtained carboxylic acid functionized
Before the preparation of the epoxy based nanoparticles the GAgNPs + CNTs was dissolved in ethanol and was degassed to reduced air in the formulation and remaining ethanol at 75°C, 4hrs.

A white Epoxy resin LY556 (HERENBA BRAND) of density 1.15-120g/cm³ and Hardener (HY951) of density 0.98 g/cm³ with structure as displayed in Figure 2b was used in the production of the composites. The matrix is prepared by mixing of unsaturated epoxy as well as hardener araldite in ratio:10:1. The mixture was stirred using a magnetic stirrer for a period of 10–15 min. A modified solution-stir-cast with roller press method was used in the production of the hybrid composites epoxy-GAgNPs+CNT by varying the % of CNTs from 0.1, 0.2, 0.3, 0.4 and 0.5 using a constant 0.5%GAgNPs. During the production a roller pressure was used to remove air bubbles and to ensure homogeneity. The produced sample was then cast into a wooden mold, cured at 75°C for 4hrs and post-cured at 120°C for 2hrs. The morphology and particle size of the CNTs, AgNPs, and blended GAg.NPs+CNTs were determined by transmission electron microscopy (TEM) using a model Jeol JEM-2100F. UV-2450 Shimadzu UV spectrophotometer was used to determine the absorption spectra of the biosynthesis GAgNPs. The produced composites are displayed in Figure 2c.

The electrical conductivity was conducted using the Kaise insulation test (model SK5010), the test was determined by placing the samples between two electrodes. The electrical conductivity was computed from Equation 1.

\[ \sigma = \frac{1}{\rho} = \frac{d}{(R_p)A} \]  

(1)

where: \( A \) is the area, \( d \) the thickness, \( C_p \) the capacitance, \( \rho \) = electrical resistivity.

A VEGA 3 TESCAN model scanning electron microscope(SEM) was used to determine the various microstructure obtained, X’Pert Pro model diffractometer(XRD) was used for the identification of the different phases. A testometric universal testing machine was used to determine the tensile strength. The tensile test was determined as per ASTM D3039 The thermogravimetric machine model(STA 6000) was used for the thermal analysis. The test was conducted as per ASTM E1131 with a heating rate of 20°C/min. The dynamic mechanical test was done with a dynamic mechanical machine model (Perkin Elmer 8000). The test was conducted as per ASTM D4065 using a heating rate of 10°C/min and 5Hz.

### 3.0 Results And Discussion

#### 3.1 UV absorption spectra and TEM Analysis of the GAgNPs

The biosynthesized AgNPs color changes from colourless to yellow and them yellow brown confirmed the synthesis of the GAgNPs(Figure 2a). The surface Plasmon resonance obtained by the Cashew leaves was around 450nm using UV–Vis Spectra(Figure 3a), this confirmed that GAgNPs was produced. The
broad spectra indicate the presence of particles with a broad size distribution as results of the reduction reaction of Ag\(^+\) ion into Ag nanoparticles[29].

TEM analysis was used to determine the size and shape of the GAg.NPs. The result of the TEM image of the synthesized GAg.NPs is displayed in Figure 3b. It was observed that round and spherical shapes were visible. This microstructure was well distributed without any cluster of particles. The estimated particle size of the GAgNPs was 45nm from the ranges of 5 to 100nm. The size obtained of the synthesis GAgNPs are in par with the studied reported by[29-30]

The EDS shows higher peaks of the Ag element. This confirms the high Ag signal and the formation of GAg.NPs. The purity of the GAgNPs produced was high as there is not any oxygen or silver compound formed as displayed by the EDS. This was attributed to the fact that the reduction was obtained. A similar observation was obtained in the work of [30].

3.2 SEM analysis of the samples

The SEM images of the composites are displayed in Figure 4. It is clear from these images that there was quite a difference in the morphology between the samples. Figure 4a shows the microstructure of the neat epoxy polymer. However, the addition of 0.2CNTs + 0.5%GAg.NPs and 05CNTs+ 0.5%GAg.NPs altered the epoxy microstructure (see Figure 4b and 4c). As the amount of GAgNPs and CNTs increased in the epoxy, the amount of GAgNPs and CNTs become clearly visible in the SEM. The presence of GAgNPs and CNTs can be seen clearly as white dots in the epoxy matrix. These white phases may raise the free electrons and improve the polymer conductive, energy storage ability and strengthening the polymer.

3.3 XRD analysis

The XRD spectrum obtained is displayed in Figure 5 where it was observed that the GAgNPs were stronger attached to the CNTs. The proper bonding obtained is attributed to the functionalization of the CNTs. There was a significant change in the XRD patterns of all the samples in question. The XRD pattern of the epoxy polymer shows amorphous phases which are clear features of polymer materials [31-32]. Crystalline phases were observed in the GAgNPs and CNTs. The graphite crystalline phases in the CNTs as revealed by the XRD analysis are: (100), (102), and (110) respectively. This is on par with the XRD analysis observed by [33-34]. The XRD patterns of GAgNPs revealed silver metallic phases of (111), (200), (220), and (311) respectively.

The addition of 0.5% GAgNPs and 0.5%CNTs to the epoxy polymer altered the amorphous nature of the epoxy polymer to the amorphous-crystalline phase The presence of GAgNPs and CNTs were the main reason for the crystalline phases of GAgNPs (111 and 200) and CNTs(100) were observed in the XRD spectrum(see Figure 5). The binding energy between GAgNPs and CNTs observed in this work were attributed to the functionalization of the CNTs which improved the bonding and reactivity of the GAgNPs [34]. The GAgNPs produced more diffraction peaks in all the samples under investigation. Equation 2 was
used to determine the crystalline size of the GAgNPs, CNTs, and epoxy-GAgNPs+CNTs. It was observed that a crystalline size of 7.56nm, 10.45nm, and 15.56nm were observed for the GAgNPs, CNTs, and epoxy-GAgNPs+CNTs respectively.

\[ D = \frac{0.9 \lambda}{\beta \cos \theta} \]  
\[ (2) \]

3.4 Electrical Conductivity.

The addition of CNTs and GAgNPs increased the electrical conductivity of the epoxy. It was observed that an electrical conductivity of \(5.6 \times 10^{-13} \text{S/cm}\) was obtained for the pure epoxy, this value is within ranges reported in the literature for epoxy [35]. The addition of CNTs and GAg decorated CNT has a great effect on electrical conductivity. It can be observed in this work that both epoxy/CNTs and epoxy/GAg+CNTs composites resulted in an increase electrical conductivity (see Figure 6), however the epoxy/GAg+CNT show a higher increment in the electrical conductivity than the epoxy-CNT composites. The increment in the electrical conductivity of epoxy/GAg+CNTs than epoxy-CNT was attributed to the higher electrical conductivity of AgNPs [36] than that of the CNTs. The electrical conductivity of the epoxy polymer has been enhanced from \(5.6 \times 10^{-13} \text{S/cm}\) to \(4.80 \times 10^{-3} \text{S/cm}\) for epoxy/0.5%CNTs and \(9.1 \times 10^{-3} \text{S/cm}\) epoxy/0.5%CNTs +0.5%GAgNPs respectively. The higher electrical conductivity observed can be also attributed to the higher surface area to volume of the GAgNPs which helps to create a conductive pathways in the polymer resin[37].

3.5 Tensile Strength

Tensile strength is one of the most important features of a conducting polymer. The ability of polymer materials used in electronic devices to withstand high strength when used as a conducting polymer is very vital. Figure 7 displayed the stress-strain curves of the epoxy, epoxy-0.5%CNTs, and epoxy-0.5%CNTs+0.5%GAgNPs. The 0.5% GAgNPs was chosen for the tensile strength analysis because the values have higher electrical conductivity. The epoxy-0.5%CNTs+0.5%GAgNPs have a higher region under the stress-strain curve with equivalent strain of 0.049mm/mm, while that of epoxy-0.5%CNTs was 0.038mm/mm and 0.026mm/mm for the epoxy respectively. The raise in the stress-strain curve of the epoxy-0.5%CNTs+0.5%GAgNPs means it is the toughest among the sample under investigation.

However, the addition of CNTs and GAgNPs to the epoxy polymer enhanced the tensile strength. For example, elastic modulus of 1.31, 2.45, 2.58, and tensile strength of 47.5, 71.2, 80.00MPa were obtained for the epoxy, epoxy-0.5%CNTs, and epoxy-0.5%CNTs+0.5%GAgNPs respectively. The additions of CNTs and GAgNPs epoxy help to improve the stiffness of the epoxy and alter the polymer network. The improvement of the tensile strength of the epoxy-0.5%CNTs+0.5%GAgNPs than epoxy-0.5%CNTs was attributed to the higher toughness of the GAgNPs than the epoxy and CNTs, help to decrease the brittle
nature of the epoxy and closed the polymer network bridge and improved the strength. The GAgNPs can be effective used to improve the strength of conducting epoxy-CNTs for electronic devices.

3.6 Thermal Conductivity

Thermal conductivity is an important factor of polymer used in electronic devices. The GAgNPs was used to enhance the thermal conductivity of epoxy-CNTs. The thermal conductivity of the epoxy was significant enhanced with the increase in the addition of CNTs in the formulation. The rise in the thermal conductivity of the epoxy-CNTs was attributed to the high thermal conductivity of the CNTs(740) than epoxy(0.48)(see Figure 8). The thermal conductivity of the polymer has been enhanced from 0.48 W(m/K) to 1.02W(m/K) and 1.23 W(m/K) for epoxy-0.5%CNTs and epoxy-0.5%CNTs+0.5%GAgNPs respectively.

It was observed that the thermal conductivity of the epoxy-GAgNPs+CNTs is quite higher than the thermal conductivity of the epoxy-CNTs. The rise in the thermal conductivity of the epoxy-GAgNPs+CNTs was attributed to the high thermal conductivity of the AgNPs. The GAgNPs help to improve the conductance of the CNTs, the matrix network, enhanced phonon conduction and decreased boundary scattering [38]. It was established that GAgNPs can be promising materials to the enhanced thermal conductivity of epoxy-CNTs and recover the potential reduction.

3.7 Thermal Analysis

Thermogravimetric analysis (TGA) is an important method used to determine the thermal analysis of materials. TGA gives the weight loss of materials as a function of temperature and degradation temperature. It was observed that the composite with epoxy-0.5%CNTs+0.5%GAgNPs has better thermal resistance than the epoxy, epoxy-0.5%CNTs composite respectively. The epoxy-0.5%CNTs+0.5%AgNPs have a higher residual mass retained as displayed in Figure 9. The higher thermal resistance of the epoxy-0.5%CNTs-0.5%GAg-NPs can be attributed to the formation of silver oxide. The silver oxide formed delayed the rate of burning and enhanced the thermal efficiency of the composite for an electronic application. A similar observation was obtained in the work of [4].

The materials in question exhibit single DTG at temperatures between 300-500°C(Figure 10). Maximum temperatures of degradation occurred between 350-400°C. However, the DTG of the composites shifted to a higher temperature due to the thermal stability of the epoxy-0.5%CNTs+0.5%AgNPs. The low peak observed at temperatures between 100-200°C was attributed to the evaporation of water in the samples. The epoxy-0.5%CNTs+0.5%GAgNPs has the highest temperature of maximum degradation of all the samples under investigation.

3.8 Dynamic Mechanical Strength

The results of the dynamic mechanical properties are displayed in Figure 11. The storage modulus displayed the information such as the interfacial bonding, elasticity of the materials, and stiffness with
temperature. It displayed the glassy, transition, and rubbery regions in the materials. It can be seen that there was an increase in the storage modulus at the frozen stage of the glassy temperature. It was observed that the decrease in the values of the storage modulus observed above the glassy transition temperatures was as a result of the ease of movement of the polymer chain as temperature increases.

The composites developed from epoxy-0.5%CNTs and epoxy-0.5%CNTs+0.5%GAgNPs have a higher storage modulus in both the glassy and rubbery than the epoxy polymer (see Figure 11). This great increment in the values of storage modulus in the elastomeric and glassy ranges of the samples; this phenomenon can be explained to the fact that the strength obtained in this region depend on the package of the polymer chain and intermolecular forces that hold the polymer chain together [36-37]. However the reduction in the storage modulus was lower in the composites as compared with the epoxy matrix, Effective restriction of extensive molecular motions of the matrix results in the better reinforcement, restricting the plastic deformation of the polymer matrix [5].

It was observed that the addition of CNTs and GAgNPs to epoxy increases the glass transition temperature. The epoxy-0.5%CNTs+0.5%GAgNPs have a higher Tg of the entire sample. The higher Tg of epoxy-5%CNTs-5%GAgNPs composite can be attributed to good interactions between CNTs and GAgNPs leading to the formation of good CNTs and GAgNPs networks and restricted the ease of movement of the epoxy polymer chains. The increase in the storage modulus of the composites could be attributed to addition of GAgNPs which increase in stiffness of the matrix, permitted a better degree of stress transfer at the interface and blocking the interlocking molecular chains of the polymer, this reduced the ease mobility of the polymer chains under load as temperature increases [36-37].

### 4.0 Conclusions

GAgNPs was successful used in the production of epoxy-CNTs hybrid composites. The uniform distribution of CNTs and GAgNPs in the epoxy matrix formed strong particle-epoxy phases. The electrical conductivity of the epoxy polymer has been enhanced from $5.6 \times 10^{-13} \text{S/cm}$ to $4.80 \times 10^{-3} \text{S/cm}$ for epoxy-0.5%CNTs and $9.1 \times 10^{-3} \text{S/cm}$ epoxy-0.5%CNTs-0.5%GAg-NPs. The GAg-NPs can be effective used to improve the strength of conducting epoxy-CNTs for electronic devices. The addition of CNTs and GAgNPs to epoxy increases the glass transition temperature. DTG of the composites is shifted to higher temperature due to the thermal stability of the epoxy-0.5%CNTs-0.5%Ag.NPs. It was established that GAgNPs can be promising materials to enhanced thermal conductivity, strength, electrical conductivity of epoxy-CNTs, and recover the potential reduction for electronic devices application.

### Declarations

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Ethical Approval

This work does not include human and animal, hence does not required ethical approval from any committee.

Consent to Participate

This work does not include human and animal hence does not required Consent to participate in the research.

Consent to Publish

The Authors give the publisher the consent to publish the work.

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There is no competing of interests to report in this work

Availability of data and materials

The authors confirm that the data supporting the findings of this study are available within the article

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