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Characterization and Electrical Performance of Epoxy-Filled Graphite and Carbon Black Conductive Ink via Ball Milling on Polyethylene Terephthalate

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Conductive ink printing is a novel approach in the electronics industry, offering notable electrical conductivity. Epoxy serves as the binder due to its compatibility with various curing agents and fillers, as well as its excellent adhesion. Graphite (G) and carbon black (CB) are ideal conductive fillers, enhancing both electrical conductivity and adhesive strength. Their synergistic interaction forms a continuous network that stabilizes electrical properties. This research focused on the effects of different G-CB ratios in epoxy-based conductive ink on electrical conductivity and adhesion when applied to a polyethylene terephthalate (PET) substrate. FTIR was used to detect functional groups, while FESEM examined the filler dispersion in the epoxy matrix. Conductivity was measured using a multimeter following ASTM F390 standards, and the Four-Point Probe (4PP) technique was applied according to ASTM F84. The optimal G-CB ratio of 1:2 yielded the best results, with a multimeter reading of 1.71×10^{-3} S/m and a 4PP reading of 3.44×10^{-3} S/m. Adhesion, assessed via a cross-cut test, also showed strong performance at this ratio, with no ink detachment. These results highlight the potential of this G-CB conductive ink formulation for printed electronics applications.

Keywords: Conductive ink; epoxy; graphite; carbon black; polyethylene terephthalate

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Conductive ink has gained popularity due to its adaptability in a variety of applications, such as flexible electronics, RFID tags, membrane switches, and touchscreens, as it allows current to flow through it. There are many different types of conductive inks in the market, and selecting the right one for electrical devices is crucial [1]. Additionally, conductive ink is particularly versatile since it can be applied to a variety of substrates, including flexible and curved surfaces, making it perfect for flexible electronics [2]. Carbon-based conductive inks are less expensive and more biocompatible than other similar inks, which has garnered interest in studies for the fabrication of various flexible electronic devices [3]. Their biocompatibility ensures safe usage in medical and wearable electronics, such as biosensors, drug-delivery devices, and tissue-engineering scaffolds, without causing adverse reactions [4]. Binder matrix, conductive filler, and solvent are the three major components of conductive ink. A typical binder in conductive ink compositions is epoxy resin, due to its excellent adhesive and mechanical strength, as well as its adaptability in terms of curing agent and filler options. However, epoxy cannot conduct electricity by itself. Hence, a carbon-based filler, such as graphite (G), was used as a filler in this study due to its great electrical conductivity, affordability, ecologically sustainable nature, and availability, qualities which make it highly desirable for the manufacture of printed electronics. G is a naturally conductive material due to its structure, with layers consisting of graphene sheets stacked on top of each other, allowing the movement of electrons. It is more cost-effective compared to some other conductive materials like gold or graphene. Apart from that, G is abundant and readily available, contributing to its affordability, and it can be easily dispersed in solvents or binders to form a stable ink [5]. Proper dispersion is crucial to ensure a well-dispersed and stable ink, since uniform dispersion of nanoparticles inside the resin matrix is a key difficulty when employing resins as a binder in conductive inks. Although G is recognized as an excellent conductor of electricity, its practical use is limited by its tendency to agglomerate at higher concentrations due to strong self-interactions [6]. Restacking happens regularly during mixing due to weak intermolecular interactions between graphene layers and the polymer matrix. Thus, it is essential to improve the dispersion of G in order to overcome the following problem.

Several efforts have been made to develop hybrid inks by integrating graphite with metal nanoparticles or other conductive polymers. According to recent research, using a mixture of different carbon fillers could create a synergistic effect, offering a viable alternative for enhancing electrical conductivity while improving the overall performance of conductive inks. Electrical conductivity likely arises from the combination of two conductive fillers with distinct shapes and aspect ratios, which leads to the formation of continuous network structures within the polymer [7]. The synergistic effect is crucial, as it demonstrates that the fillers are well combined and evenly dispersed, allowing them to be packed more closely together. This approach not only prevents agglomeration but also helps reduce material costs. Among of all the carbon materials, carbon black (CB) was selected for its affordability, large surface area, and excellent electrical conductivity [8]. Recent research shows that adding carbon nanoparticles, such as G or CB, to polymer composites can create a synergistic effect, leading to improved mechanical properties, as well as enhanced thermal and electrical conductivity and piezoresistive behavior. Using a mixture of different carbon fillers is a promising approach to balance performance and cost. By incorporating carbon fillers with varying aspect ratios and sizes into a polymer matrix, signs of exfoliation and particle size reduction can be observed during the mixing process, including when expanded graphite is used. This suggests that mechanical milling can help exfoliate graphite layers into smaller dimensions [9].

CB consists of small particles, typically with diameters in the nanometer range, a high surface area, and an approximately spherical shape. Since most polymers are naturally non-conductive and do not allow the flow of electricity, in some applications, adding CB to the material's matrix might result in the required performance, such as conductivity. Depending on the chemistry used and the physical characteristics of the material, the resistivity values of CB typically range from 10^{-1} to 10^{12} cm [10]. According to a study by Mailabari et al., the resistivity of CB that is combined with epoxy resin is affected by the morphology, size of nanoparticles, and loading content of the CB [11]. Another study by Philips et al. discovered that combining carbon black with graphite as fillers in a polymer binder creates a much more conductive matrix than using each material on its own. The small carbon black particles disperse well and form conductive links between graphite-rich regions within the composite. Their study showed that the screen-printed ink achieved optimal conductivity, measuring 0.029 Ωcm, when a ratio of 2.6:1 of graphite: carbon black was used [5]. CB functions as a coupling agent, enhancing the overall properties of the ink, while G particles serve as spacers that keep CB aggregates apart, preventing them from clustering together into larger groups [12]. High-speed mixing helps reduce the size of CB particles during their refinement. During this process, G particles can coat the CB particles, leading to various interactions like electrostatic forces, hydrogen bonding, and van der Waals forces.

The study of adhesion between conductive ink and plastic substrates is crucial for evaluating the performance and reliability of printed electronic devices. Conductive inks are commonly used in flexible electronics, RFID, sensors, and other applications where they are printed or applied to plastic surfaces. Strong adhesion is important for maintaining the integrity, functionality, and resistance of the conductive patterns against environmental factors and mechanical stress. Poor adhesion can result in issues such as peeling, cracking or reduced electrical performance, ultimately affecting the device's efficiency and durability. By studying adhesion properties, researchers can identify ideal ink formulations, surface treatments or compatibility improvements that enhance bond strength and stability between the ink and the substrate, leading to better device performance [13]. Different plastic substrates have distinct adhesion characteristics, with polyethylene terephthalate (PET) being commonly used due to its widespread application as an engineering plastic, especially when combined with materials like carbon nanotubes and glass fibers to enhance its strength. PET, a thermoplastic polymer resin, is widely utilized in packaging, textiles, films, automotive, electronics, and many other industries, making it a suitable candidate for this research [14].

The aim of this study is to develop a conductive ink by optimizing the ratios of G and CB in an epoxy matrix, minimizing agglomeration while achieving high electrical conductivity. Different ratios of hybrid Epoxy-G-CB (0:0, 0:1, 1:0, 1:2, 4:1) were synthesized using a green cosolvent method to evaluate their synergistic effect on characterization properties and conductivity levels. To study the adhesion properties of the conductive ink on the PET substrate, the cross-cut method was used. Additionally, this research aims to address the gap in existing studies by investigating the interactions between different fillers within the resin matrix, which serves as the motivation for this project.

EXPERIMENTAL

Chemicals and Materials

The study employed Bisphenol-A-(epichlorohydrin) epoxy resin, with 3-aminomethyl-3,5,5-trimethylcyclohexylamine as the curing agent. Carbon black N330, with a particle size of approximately 28-36 nm, and graphite, with a particle size of less than 20 µm, both sourced from Sigma Aldrich, were used as fillers. Sodium dodecyl sulfate (SDS), with a particle size of around 110 µm and sourced from R&M Malaysia, was utilized as a surfactant. Ethanol and distilled water were also part of the formulation. The equipment and instruments used in the analyses included Crest Ultrasonic Model 4HT-1014-6 ultrasonic bath, a centrifuge, a laboratory planetary ball mill, Nichiban tape, PET film as the substrate, Perkin Elmer Spectrum RX1 FTIR, Supra 55VP FESEM, a digital multimeter, and Keithley 6487 picoammeter connected to 4PP.

Preparation of Hybrid Epoxy-G-CB Conductive Ink

Different G-CB inks were prepared at five distinct ratios: 0:0, 1:0, 0:1, 1:2, and 4:1. Each mixture was dispersed in a cosolvent of ethanol and distilled water, combined in a 2:1 ratio, along with 0.1 wt% of SDS surfactant. The dispersion was sonicated for about one hour, followed by ball milling in a laboratory planetary mill for another hour at a rotation speed of 100 rpm. After ball milling, the resulting mixture was centrifuged three times at 6000 rpm for 15 minutes to obtain a paste containing suspended G and CB particles. Subsequently, 2 mL of Bisphenol-A-(epichlorohydrin) epoxy resin was added to the paste, and the mixture was further processed by ball milling for 3 hours at 100 rpm. Finally, 1 mL of the curing agent (3-aminomethyl-3,5,5-trimethylcyclohexylamine) was added, and the mixture was screen-printed onto a PET film.

Characterization Studies

To confirm the presence of G and CB in the epoxy resin matrix, FTIR analysis was used to identify the functional groups in the prepared samples. The evaluation was performed in thin film form, with a resolution of 4 cm^{-1}, over a spectral range of 400 to 4000 cm^{-1}. FESEM was employed to observe the morphological structure of the conductive ink, focusing on its dispersion behavior and potential agglomeration. A cross-section of the thin conductive film was examined, which had been cut using nitrogen gas and coated with gold sputter for enhanced imaging quality.

Conductivity Test

A digital multimeter was utilized to assess the conductivity level of the conductive ink printed on the PET film, following the standard procedure outlined in ASTM F390, which is commonly used for thin films. The multimeter provided a live reading of the resistance value, which was then converted into conductivity using Equation (1).

$$\sigma = \frac{A}{t \times R} \tag{1}$$

Where,

R = Resistance reading by the multimeter (Ω)
A = Cross-sectional area of the conductive film (m^2)
t = Thickness of the conductive film (m)
σ = Conductivity (S/m)

A four-point probe (4PP) was performed in accordance with ASTM F84, a method commonly used to measure the sheet resistance of thin films. This technique, combined with computer automation, allows for quick and automated testing of sheet resistance readings, which are subsequently calculated using Equations (2), (3), and (4) to obtain the conductivity values.

$$Rs = \frac{\pi}{\ln(2)} \frac{\Delta V}{I} = 4.53236 \frac{\Delta V}{I} \tag{2}$$

$$\rho = Rs \times t \tag{3}$$

$$\sigma = \frac{1}{\rho} \tag{4}$$

Where,

ΔV = Change in voltage measurement between the inner probes
I = Current applied between the outer probes
ρ = Resistivity (Ω·m)
Rs = Sheet resistance (Ω/sq).
t = Thickness (m)
σ = Conductivity (S/m)

Adhesion Test

To evaluate the adhesion of printed ink to a PET film substrate, the cross-cut method according to ISO 2409 standards was employed. A grid was cut into the ink using a multi-blade tool, after which an adhesive tape was applied to the grid and subsequently removed. This process revealed the level of adhesion based on the amount of ink that flaked off onto the tape. The assessment was visually evaluated on a scale from 0 to 5, indicating the degree of ink adhesion.

RESULTS AND DISCUSSION

FTIR Analysis

The functional group properties for various G-CB ratios in the epoxy matrix are compared in Figure 1. The spectrum for the epoxy G-CB 0:0, representing pure epoxy, shows the typical absorption bands associated with Bisphenol-A-(epichlorohydrin) epoxy resin. A distinct absorption band between 3600 and 3200 cm^{-1} corresponds to the OH bond, while a weak band at 3664 cm^{-1} represents the stretching vibrations of "free" O-H groups. However, this peak is small due to the limited number of OH groups in the resin's molecular structure [15]. The 950 cm^{-1} band signifies the presence of epoxy groups, and the peak at 1100 cm^{-1} indicates the presence of aromatic ether (C-O-C) groups from the epoxy ring. Importantly, no residual absorption at 3380 cm^{-1} (NH_2) is observed in the neat epoxy spectrum, confirming that all the amine groups have fully reacted during the curing process [16].

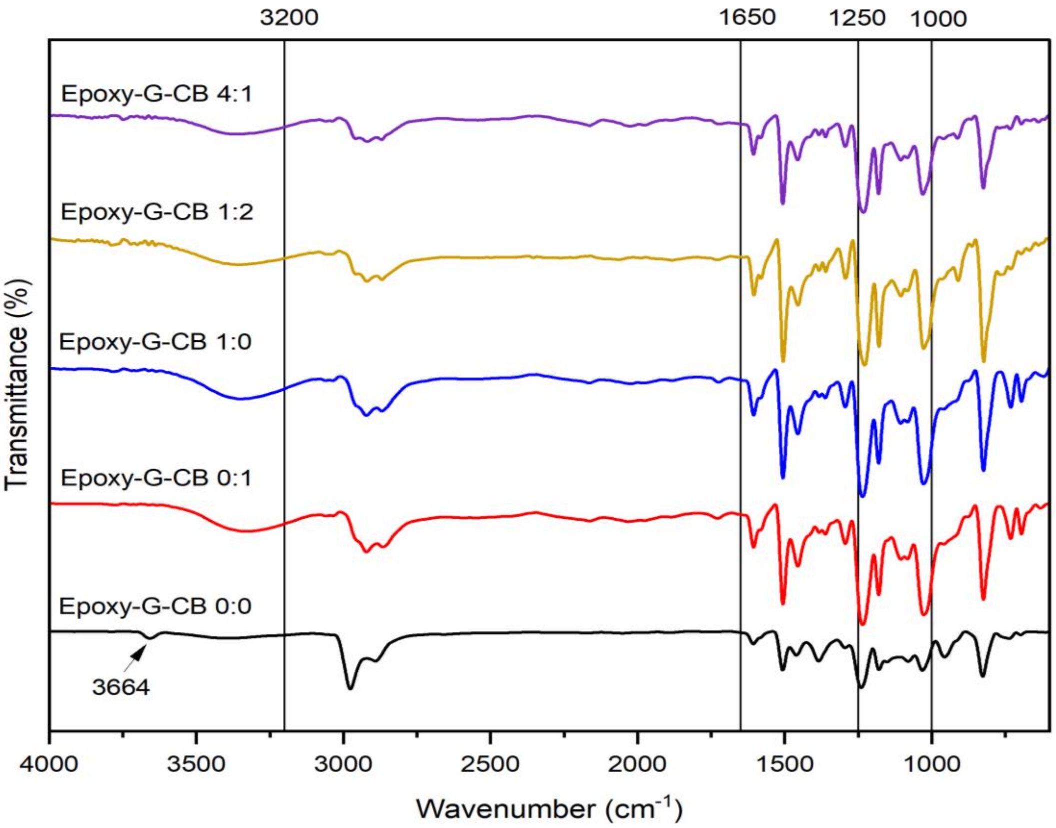


Figure 1. FTIR spectra of different ratios of Epoxy-G-CB.

The spectra obtained for epoxy containing G and CB fillers exhibit a broad OH band between 3600 and 3200 cm^{-1}, indicating that amine-cured epoxy resins primarily form hydrogen bonds with the surfaces of carbon fillers. These interactions occur between the polymer's amino or epoxy groups and the carboxyl or hydroxyl groups of the fillers. According to Horie et al., the interface between the filler and the resin suggests potential molecular structures for both hydrogen and covalent bonds [17]. In the FTIR spectra of epoxy G-CB ratios of 0:1, 1:0, 1:2, and 4:1, hydroxyl groups are observed in the band between 3200 cm^{-1} and 3500 cm^{-1}, while the vibration associated with carboxyl group formation appears at 1241 cm^{-1}, further supporting the formation of these bonds. The spectra for G-CB 0:1 and 1:0 correspond to the carbon black and graphite incorporated in the epoxy resin, respectively. Meanwhile, the G-CB 1:2 and 4:1 mixture represent a combination of the spectra of epoxy, G, and CB. In the epoxy-G-CB 1:2 sample, the intensity of the carboxyl group is the highest, indicating a greater formation of covalent bonds compared to the other samples. However, no new peaks appear with the addition of CB and G fillers, although the intensity of the characteristic composite peaks increases with higher filler concentrations. The consistent presence of these peaks across all G-CB samples indicates successful dispersion of the fillers within the epoxy matrix. This effective dispersion enhances the overall conductivity of the ink by creating a conductive network. A typical band corresponding to the aromatic C=C bond stretching vibrations of an aromatic ring is observed in the 1600–1400 cm^{-1} range. In the G:CB ratio of 4:1, the peaks for CB are less noticeable, likely because they are overshadowed by the layered structure of G.

Surface Fracture Morphology Studies

The FESEM cross-sectional images of fractured conductive film samples at various ratios are shown in Figure 2. Assessing the morphology of the fracture surface is essential for understanding the internal distribution of conductive particles within the epoxy matrix. In Figure 2(a), the fracture surface of the neat epoxy, without any filler particles, appears smooth, which is characteristic of epoxy resin morphology. This smooth fracture surface indicates the brittle nature of the neat epoxy, demonstrating poor fracture toughness due to its low resistance to crack initiation and propagation under load [18]. Comparing Figure 2(a) to the other images, it is evident that the addition of fillers to the epoxy matrix improves its fracture toughness.

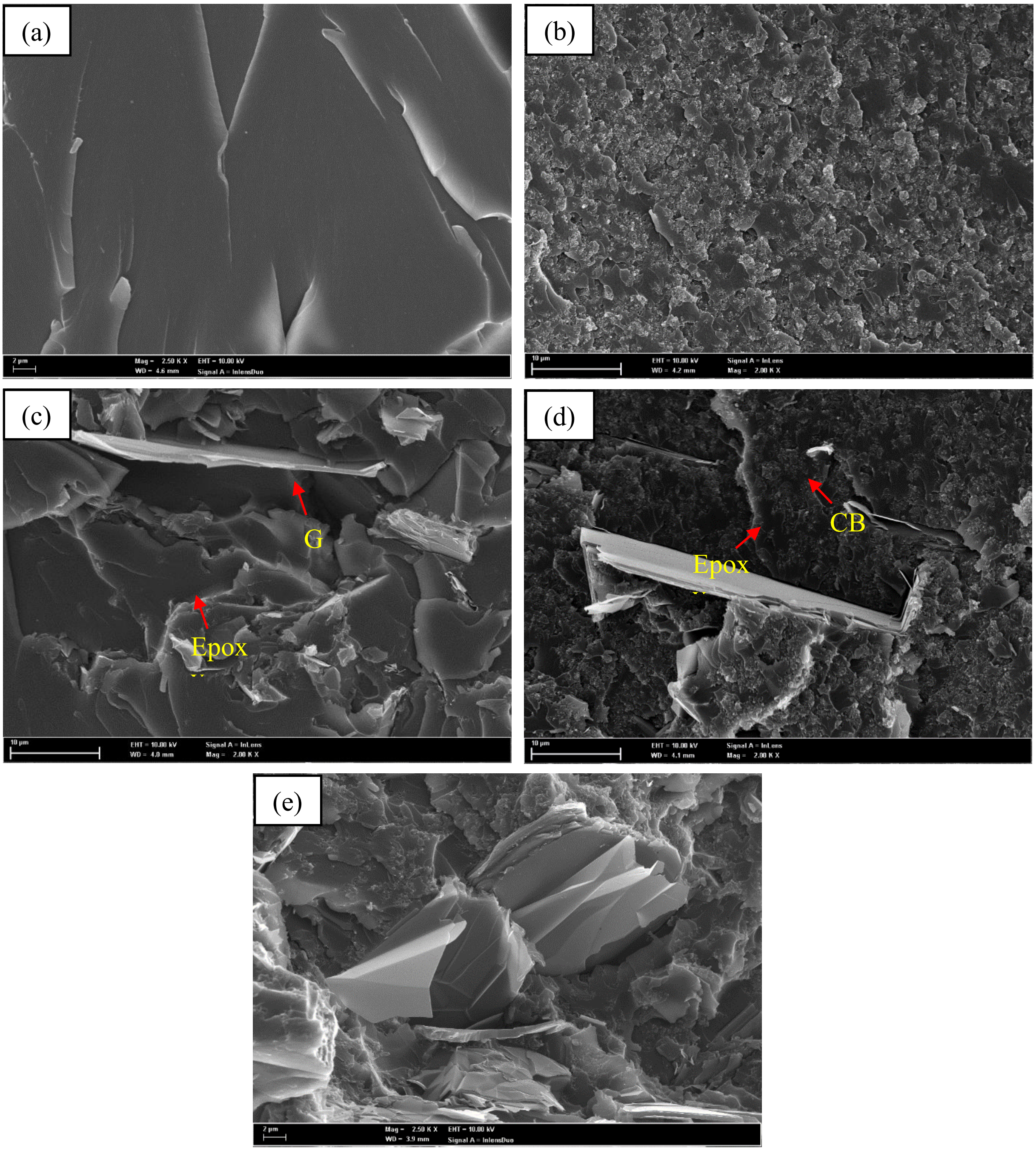


Figure 2. FESEM micrographs of conductive inks: (a) Epoxy-G-CB 0:0; (b) Epoxy-G-CB 0:1; (c) Epoxy-G-CB 1:0; (d) Epoxy-G-CB 1:2; and (e) Epoxy-G-CB 4:1, at 2000X magnification.

As depicted in Figure 2(b), the FESEM image of the Epoxy-G-CB at a 0:1 ratio shows small particle aggregates dispersed within the epoxy matrix, likely in the form of spherical CB particles. At this ratio, G-CB demonstrates a fairly uniform dispersion of CB particles. The smaller CB aggregates contribute to more efficient distribution, increasing the contact area between CB and epoxy. This leads to closely interconnected particles, creating bridging networks that facilitate easier electron transport, thus enhancing conductivity [19]. However, the CB particles seem to overlap, which may hinder the formation of additional conductive pathways compared to the G-CB 1:2 ratio. Meanwhile, in the G-CB 1:0 ratio, a flake-like structure with uneven formation is observed, likely due to the G particles. Figure 2(c) illustrates that the G flakes are embedded within and adhered to the epoxy matrix, rather than stacking with each other [20]. However, the dispersion of G particles within the polymer matrix is poor, leading to agglomeration. This is caused by weak interactions between the matrix and the filler. The insufficient bonding between epoxy and the G filler contributes to this agglomeration.

A noticeable improvement in the dispersion of G-CB is observed in the G-CB 1:2 ratio in Figure 2(d). In this case, both the flake-like micro-G structures and spherical CB particles are evenly distributed within the epoxy matrix. The G particles act as spacers between the CB particles, forming a 3D "point-surface-point" conductive micro-nanostructure network. This arrangement enhances the interaction among the conductive fillers, resulting in more conductive pathways and thus improving the overall conductivity of the material [21]. In contrast, at the 4:1 ratio, as shown in Figure 2(e), the G platelets tend to stack on top of each other, leading to aggregation within the epoxy resin rather than uniform dispersion, which impedes electron transfer and results in lower conductivity [22].

Multimeter

Table 1 and Figure 3 display the multimeter readings and the relationship between resistivity and conductivity. An inverse correlation is observed, indicating that as resistivity decreases, conductivity tends to increase. Resistivity indicates the resistance to the flow of electricity, while conductivity indicates the ability to conduct electrical current. The resistance reading for the G-CB 0:0 sample, which contained no filler, is exceptionally low, shown as "OL" on the multimeter. This means the resistance is beyond the measurement range, as epoxy resin is an insulator with resistance in the megaohm or gigaohm range. However, when fillers were incorporated in the G-CB 0:1 and 1:0 samples, the multimeter displayed resistance values that correspond to calculated conductivities of 7.34×10^{-5} S/m and 4.49×10^{-5} S/m, respectively. This improvement is attributed to the significant increase in conductivity caused by the filler addition. The fillers form a conductive network within the epoxy, creating continuous pathways for electron movement and enhancing the composite's overall conductivity [23].

The optimum ratio for G-CB is found to be 1:2, resulting in a significant reduction in resistivity and yielding the best electrical performance. As shown in Figure 3, the highest conductivity of $1.71 \text{ x } 10^{-3}$ S/m is achieved at the 1:2 ratio, which corresponds to the lowest resistivity of $5.85 \text{ x } 10^{2}$ Ωm. This behavior, as noted in the literature, is attributed to the incorporation of small CB particles into polymer-G composites, which fills the gaps between larger G particles and creates new conductive pathways [24]. Additionally, the smaller size of CB allows for better dispersion within the matrix, minimizing insulating regions in the material. The G particles act as spacers, keeping CB aggregates apart and preventing them from forming larger clumps [12]. However, at higher filler loadings, an excess of the hybrid filler can cause aggregation, which reduces electrochemical performance and diminishes the synergistic effects [25]. This explains why the conductivity of G-CB at the 4:1 ratio decreases to $3.57 \text{ x } 10^{-5}$ S/m, with a resistivity of $2.80 \text{ x } 10^{4}$ Ωm. Agglomeration is more likely to occur when graphite particles cluster at higher loadings, disrupting the continuity of the epoxy matrix. This agglomeration creates regions of lower conductivity within the composite, restricting the flow of electrical current across the material [26].

Table 1. Resistance readings, resistivity, and conductivity of Epoxy-G-CB ratios.

Formulation	Resistance (Ω)	Resistivity (Ωm)	Conductivity (S/m)
G-CB 0:0	OL	0	0
G-CB 0:1	0.78×10^{6}	$1.36 \text{ x } 10^{4}$	$7.34 \text{ x } 10^{-5}$
G-CB 1:0	1.27×10^{6}	$2.22 \text{ x } 10^{4}$	$4.49 \text{ x } 10^{-5}$
G-CB 1:2	3.35×10^{4}	$5.85 \text{ x } 10^{2}$	$1.71 \text{ x } 10^{-3}$
G-CB 4:1	1.60×10^{6}	$2.80 \text{ x } 10^{4}$	$3.57 \text{ x } 10^{-5}$

Four-Point Probe Measurement

Two-point measurements are simple and effective, but for materials with low resistivity, such as thin films and semiconductors, four-point probe measurements are preferred due to their improved accuracy [27]. The four-point probe method focuses on determining sheet resistance, which is inversely proportional to electrical conductivity. A similar trend in conductivity is seen when using a multimeter, as presented in Table 2 and Figure 4. The control sample with a G-CB ratio of 0:0 shows the highest resistivity at 5.01 x 10^9 Ωm and the lowest conductivity at 2.00 x 10^{-10} S/m. These results are consistent with Ganguli et al., who reported that epoxy resin without conductive filler acts as an insulator, with a resistivity of 1.58 x 10^8 Ωcm [28]. At low filler ratios like G-CB 0:1 and 1:0, the particles are too dispersed to form continuous conductive networks, resulting in lower conductivity values of 3.33 x 10^{-3} S/m and 3.25 x 10^{-3} S/m, respectively. The increase in conductivity is due to the addition of conductive fillers, which form interconnected networks within the epoxy matrix, facilitating electron movement and thereby improving the overall conductivity of the composite [23].

The optimal result from the four-point probe measurement, indicating good electrical conductivity, is observed at the G-CB ink ratio of 1:2, yielding a resistivity of 2.19 x 10^2 Ωm and a conductivity of 3.44 x 10^{-3} S/m. Increasing the filler content lowers the packing density and increases structural nonuniformity within the epoxy resin. The CB particles disrupt the uniform packing of G particles by filling the spaces between them, preventing the formation of a densely packed structure. This more heterogeneous filler distribution enhances conductivity [29]. Additionally, the contact interface between the G and CB conductive fillers significantly influences the conductivity level. However, beyond the 1:2 ratio, the conductive network becomes oversaturated due to excessive filler packing, causing particles to cluster and agglomerate. At the 4:1 G-CB ratio, resistivity increases to 4.25 x 10^2 Ωm, and conductivity decreases to 2.35 x 10^{-3} S/m. Therefore, determining the optimal hybrid filler ratio is critical for optimizing the ink's performance in terms of conductivity, stability, and application properties. These results also demonstrate not only an improvement in the overall accuracy of electrical measurements but also that the multimeter and four-point probe methods complement each other, capturing essential aspects of the material's conductivity.

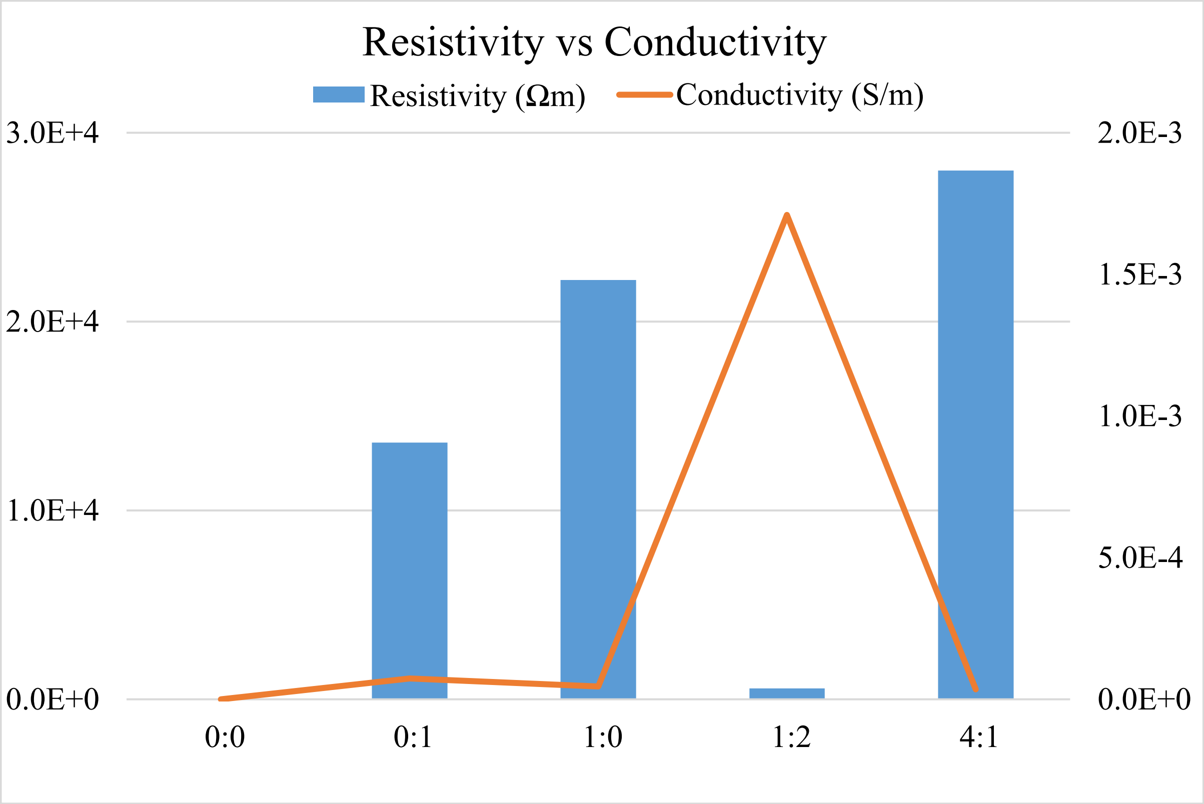


Figure 3. Resistivity vs conductivity using multimeter of Epoxy-G-CB ratios.

Table 2. Resistance readings, resistivity, and conductivity of Epoxy-G-CB ratios.

Formulation	Current, I (A)	Sheet Resistance, Rs (Ω/sq)	Resistivity (Ωm)	Conductivity (S/m)
G-CB 0:0	6.33×10^{-13}	7.16×10^{12}	5.01×10^{9}	2.00×10^{-10}
G-CB 0:1	10.56×10^{-6}	4.29×10^{5}	3.00×10^{2}	3.33×10^{-3}
G-CB 1:0	10.30×10^{-6}	4.40×10^{5}	3.08×10^{2}	3.25×10^{-3}
G-CB 1:2	10.92×10^{-6}	4.15×10^{5}	2.19×10^{2}	3.44×10^{-3}
G-CB 4:1	7.46×10^{-6}	6.08×10^{5}	4.25×10^{2}	2.35×10^{-3}

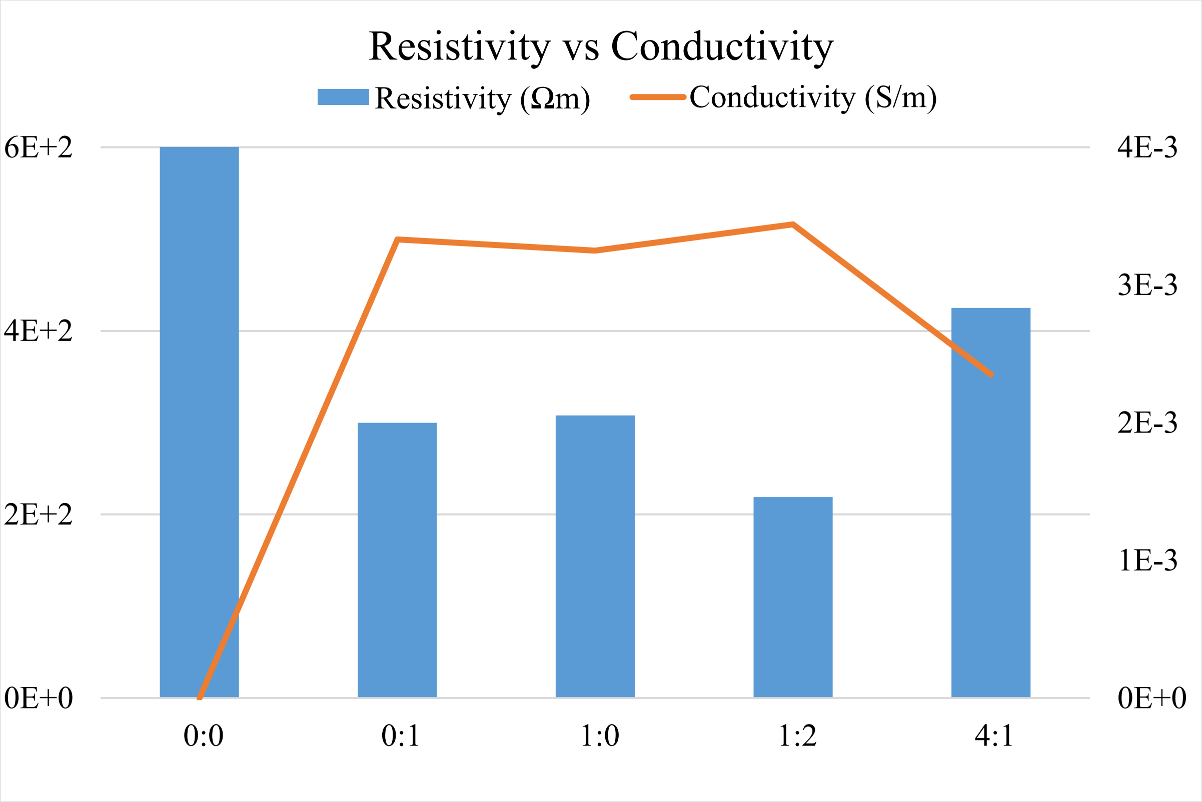


Figure 4. Resistivity vs conductivity using 4PP of Epoxy-G-CB ratios.

Cross-cut Adhesion Test

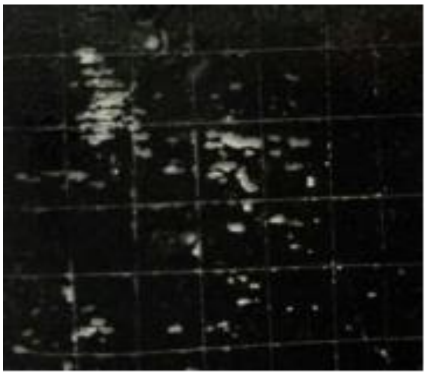
The optimal ratio of conductive fillers is essential for achieving both high electrical conductivity and strong mechanical properties in conductive inks, particularly regarding their adhesive performance on substrates. To ensure good mechanical strength, the filler loading must be optimized and evenly distributed. Various G-CB ratios underwent adhesion tests on a PET substrate, following the standard ISO 2409 cross-cut method. Adhesion loss was visually assessed and rated using the ISO 2409 scale, which ranges from 5 (indicating over 65% adhesion loss) to 0 (no adhesion loss). Table 3 shows the qualitative cross-cut test results for all samples. For the G-CB ratio of 0:0, the scale was 4, with up to 65% of the area removed, demonstrating that neat epoxy has poor adhesion to the PET substrate [30]. This is due to the lower strength of the epoxy resin compared to the increased strength provided by reinforcing the resin with fillers.

The presence of fillers is believed to enhance the adhesion between the printed ink and the substrate.

As expected, the G-CB composites at 0:1 and 1:0 ratios exhibit better adhesion properties compared to the G-CB 0:0. Specifically, the G-CB 0:1 demonstrates an area loss rating of 2 on the ASTM scale, outperforming the G-CB 1:0, which has a rating of 3, with larger ink fragments detaching from the PET substrate. This is likely due to the fact that G generally shows poor adhesion with many polymer matrices, including PET, causing adhesion problems as the G particles do not bond effectively with the PET substrate compared to the CB filler. Meanwhile, the G-CB with the 1:2 ratio displays superior adhesion, showing no area removal and a clean cut without ink fragment detachment. At the G-CB ratio of 4:1, the coverage loss is classified as 1, where only small fragments separate from the PET substrate. This improvement can be credited to graphite particles filling voids and enhancing interfacial bonding between the carbon black filler and the epoxy matrix [31], resulting in stronger adhesion between the ink and the PET substrate. The mechanical coupling or interlocking adhesion mechanism helps the conductive ink adhere and embed within surface irregularities of the substrate. Numerous studies suggest that this adhesion is primarily driven by interchain entanglement rather than chemical bonding between composite components [32].

Table 3. Cross-cut test results for Epoxy-G-CB conductive ink on PET at various G-CB ratios.

Formulation	Scale of area remove	Analysis	Surface of cross-cut area
Epoxy-G-CB 0:0	4	Some squares have become partially or completely detached, impacting between 35% and 65% of the area.	
Epoxy-G-CB 0:1	2	Small fragments of the coating have detached, affecting no more than 15% of the area.	
Epoxy-G-CB 1:0	3	The coating has detached from the square lattice, affecting 15 to 35% of the area.	

Epoxy-G-CB 1:2	0	The cuts are completely smooth, with no detachment of ink fragments	
Epoxy-G-CB 4:1	1	Small fragments of the coating have detached, affecting no more than 5% of the area.	

CONCLUSION

In conclusion, the fabricated conductive ink underwent characterization and its electrical properties were analyzed to determine the optimal ratio of filler loading. The presence of CB and G in the epoxy resin was confirmed through FTIR and FESEM analyses. FTIR results verified the interaction between the epoxy and the conductive carbon fillers, which likely occurred between the amino or epoxy groups of the polymer and the hydroxyl or carboxyl groups of the fillers. FESEM was employed to understand the internal distribution of conductive particles within the epoxy matrix. A homogenous distribution of the conductive fillers in the matrix contributed to the increased conductivity. It was observed that the G-CB ratio of 1:2 was optimal, yielding the highest conductivity of 1.71×10^{-3} S/m, as measured by a multimeter, which aligned with the four-point probe analysis, yielding a conductivity of 3.44×10^{-3} S/m. Since conductive ink is typically used in electronic devices, its adhesive properties to substrates are crucial. The cross-cut test revealed that the G-CB ratio of 1:2 exhibits superior adhesion, with no material removal and a clean cut without ink fragment detachment. Therefore, the G-CB ratio of 1:2 is determined to be the optimal loading, offering both enhanced conductivity and better adhesion compared to other ratios.

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