

Evaluation of The Adhesion Performance on GNP/Ag/SA Conductive Ink Undergoing Different Curing Time and Temperature

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Abstract

This research evaluates the adhesion performance of graphene nanoplatelet (GNP)/silver flake (AG)/silver acetate (SA) conductive ink undergoing different curing times and temperatures. The study evaluates how curing parameters, such as temperature and time, impact the resistivity and adhesion strength of the conductive ink. For the preparation of hybrid conductive ink, GNP powder was mixed with ethanol in a beaker. The GNP was sonicated in ethanol for 10 minutes. Then Ag was added to the GNP/ethanol mixture. It proceeded with the sonication for one (1) hour. At room temperature, the mixture powder was poured until it produced a fine powder. Before curing at 250°C for 1 hour, the powder was dripped with organic solvents, 1-butanol, and terpineol and mixed using a thinky mixer machine to form a paste. Next, SA was added to the mixture and sonicated for an additional hour. The solution was heated on a hotplate until the remaining ethanol evaporated. The mixed mixture was then heated in a furnace for one hour to set. The mixture was then pounded until it turned into a fine powder and dripped with 1-butanol and terpineol then mixed using a thinky mixer machine to form a paste. The hybrid conductive ink paste was then printed on copper substrates and was cured at 240°C, 250°C, and 260°C for 4, 5, and 6 hours respectively for each different curing temperature. A crosshatch adhesion test was done to assess the hybrid conductive ink's adherence to the copper substrates. In this test, the material's adhesion was evaluated using the ASTM D6677 standard. This standard uses an adhesion scale which ranges from 0B to 5B, where 5B represents the best adhesion. The findings demonstrated that the increased curing temperatures have an adverse effect on the adhesive strength of the conductive ink, highlighting the correlation between curing conditions and adhesion qualities. Future research should focus on the effects of integrating additives or modifying ink composition to improve adhesion qualities during high-temperature curing.

1. Introduction

The advancement of flexible and stretchable electronics has led to high demand for wearable electronic devices like flexible LED lights, electrodes, arrays, supercapacitors, printed patterns, photo detectors, strain sensors, and skin sensors [1]. This phenomenon stimulates extensive study in the field of conductive ink manufacture, especially focusing on improving adhesion, enhancing thermal conductivity, and electrical and mechanical performance. However, the printability is influenced by the amount of conductive filler, particle size distribution, temperature, and curing time, as well as the surface tension and energy of both the ink and substrate [2]. Thus, achieving consistent and uniform printing of conductive inks is quite challenging where the ink must be suitable for different surfaces, impacting its electrical conductivity and adhesion to the substrates [3]. Good adhesion enables materials to effectively operate as coatings on substrates [4]. Despite this, adhesion is a complex issue with various contributing factors, and the current scientific understanding of these processes is inadequate [5]. One of the contributing factors is environmental conditions, including curing temperature, which influences adhesion performance over time [6]. Comprehending the endurance and stability of adhesive bonding under different environmental stressors is crucial for determining the long-term coating performance and reliability resulting in a transition towards creating stretchable conductive ink for electronic pastes, particularly in applications requiring strong adhesion, high thermal, and excellent heat conductivity.

There has been a significant advancement in the field of graphene because of its unique physicochemical features, exceptional mechanical strength, and distinctive electrical and thermal conductivities [7]. Graphene is a single layer of carbon atoms arranged in a two-dimensional honeycomb lattice structure. The atoms are grouped in a hexagonal configuration to form a sheet of sp^2 firmly linked carbon. Graphene is recognized as the thinnest, most flexible, and strongest material known for its excellent heat and electrical conduction properties [8, 9]. Nevertheless, the electrical conductivity of pure graphene inks is often insufficient for their effective use in printed electronics applications [10]. Thus, using metallic particles in graphene-based conductive inks has gained significant interest to improve the inherent properties of graphene. Ag shows potential as a material for flexible electronics. Ag-based inks have been the primary commercial nanotechnology product and have been extensively researched globally, comparable to graphene [11]. The AgNP conductive ink is the preferred option for printed electronics due to its remarkable oxidation resistance, electrical conductivity, and other desired physical properties that allow it to adhere well to substrates [12-13]. In the realm of advanced materials for hybrid conductive ink, hybrid conductive graphene inks have been enhanced with metallic nanoparticles to increase the inherent properties of graphene and augment its functionalities across various electronic applications [12]. Hybridization of both GNP and Ag contributes to enhancements in effective thermal conductivity and heat transfer efficiency [14].

Numerous research investigations have been undertaken to investigate the effects of different curing temperatures, ranging from higher to lower, on conductive ink. The predominant results of the study suggest that the resistance of the conductive ink has an adverse relationship with temperature, particularly when subjected to higher temperatures [15, 16]. The performance of Ag ink was excellent at higher curing temperatures, with the lowest sheet resistance seen at 105°C. A contrasting pattern was seen in the case of graphite ink, whereby the resistivity exhibited a decrease during the process of low temperature curing at 50°C. The conductivity of ink was found to be enhanced at higher temperatures when it had a greater concentration of silver, whereas ink with a higher quantity of graphite demonstrated better conductivity at lower temperatures [17, 18]. To enhance conductivity, it is necessary to employ higher temperatures or prolonged annealing durations [19]. However, curing temperatures over 150°C resulted in a reduction in the resistance of conductive ink. Extended curing durations and overly elevated curing temperatures might lead to the brittleness of the conductive ink [20].

Although hybrid conductive ink is widely used in electronic applications, there is still a lack of thorough understanding of how varied curing time and temperature factors affect its adhesion ability. The most significant production parameters that influence excellent adhesion, conductivity, and mechanical properties are the substrate material's surface structure, the curing parameters (temperature and time), and the composition and cross-section area of the ink layer [21-23]. Hence, the substrate and conductive ink patterns must be resistant to bending, vibration, thermal stress, and stretching. Therefore, the utilization of a flexible substrate with printed conductive ink necessitates an impermeable microstructure, highlighting the importance of strong adhesion between the pattern and the substrate [21]. Enhancing the adhesion performance of conductive inks based on graphene is essential for the progress of printed electronics. Despite several studies that have examined how curing factors affect the electrical properties of these inks, there is still a lack of understanding about how they affect adhesion strength. Based on [40] a challenge arises from the urgent requirement for part testing and qualification methods that consider both electronic performance and functioning under adverse environmental conditions, such as high temperatures and humidity, as well as vibration and shock. This gap hinders the progress of creating dependable and long-lasting printed electronic devices. Hence, it is crucial to conduct a thorough examination of the correlation between curing time, temperature, and adhesion performance. The examination of ink adhesion to the deposition substrate can be conducted by coating adhesion tests such as scotch tape testing

and crosshatch scratch testing. However, it is important to note that these tests only offer qualitative indications of adhesion [24-26].

This research focuses on how curing parameters, such as temperature and time, impact the adhesion strength of the conductive ink using the crosshatch adhesion testing method. The purpose of this research is to examine the influence of curing parameters, specifically temperature, and duration, on the adhesive strength of conductive ink through the utilization of the crosshatch adhesion test. The aim is to gain insights into the way alterations in curing circumstances impact the ink substrate bonding process. Researchers can investigate the impact of various curing conditions on the crosslinking and solidification of ink, hence influencing its adhesion qualities, through the manipulation of curing temperature and duration. This study facilitates the optimization of the curing process in order to attain a strong and durable adhesion between the conductive ink and the substrate, hence assuring the reliability and efficacy of electrical devices or coatings.

2. Material and Methods

2.1 Material and Formulation of GNP Hybrid Conductive Ink

The main materials of highly thermal GNP hybrid conductive ink consist of GNP, Ag, SA, Ethanol, I-Butanol, and Terpeneol as shown in Table 1. GNP nanoparticle size and high aspect ratio drive the ink's electrical conductivity and mechanical strength. Ag increases electrical conductivity due to their high conductivity whereas, SA operates as a precursor for silver nanoparticles, improving conductivity and stability. As a chemical solvent, ethanol disperses filler ingredients and components, maintaining ink consistency. Additionally, it controls ink viscosity. Organic solvents like 1-butanol and terpeneol bind ink to the substrate, enhancing flow and film development. These materials work together to give the hybrid conductive ink good conductivity, adhesion, rheology, and stability for electronic applications [27]. The composition in which the material is mixed according to the weight is shown in Table 2. Based on Table 2, set in this context refers to a specific amount or quantity of composition used to formulate a conductive ink paste.

Table 1 Materials of highly thermal graphene hybridization conductive ink

Type	Material
Filler material	GNP powder 25 μ m nanoparticle size Silver flakes (Ag) Silver Acetate (SA)
Chemical Solvent	Ethanol 99.99%
Organic Solvent as binder	I-Butanol Terpeneol

Table 2 Composition of different constituents' mixture according to the weight

Material	Composition
GNP	0.5 g
Silver Flake	4.292 g
Silver Acetate	0.42 g
Ethanol	50 ml
I-Butanol	30 drops
I-Tripeneol	30 drops

For the preparation of GNP/Ag/SA hybrid conductive ink, 0.5 g of GNP powder with 25 μ m particle size was mixed with 50 ml ethanol in a beaker. The GNP was sonicated in ethanol for 10 minutes. Then the mixture was added with 4.292 g of silver flake to the GNP/ethanol mixture. It proceeded with the sonication for one (1) hour. At room temperature mixture powder was pounded until it produced a fine powder. Before curing at 250°C for 1 hour, the powder was dripped with organic solvents, 1-butanol, and terpeneol and mixed using a thinky mixer machine to form a paste. Next, 0.42 g of silver acetate (SA) was added to the mixture and sonicated for an additional hour. The solution was heated on a hotplate until the remaining ethanol evaporated. The mixed mixture was transferred to a small white porcelain beaker and then heated at 250 °C for one hour to set. Following the curing procedure, the mixture at room temperature was pounded until it turned into a fine powder. The powder was then dripped with 30 drops of 1-butanol and terpeneol and mixed using a thinky mixer machine to form a paste.

2.2 Sample Preparation

Metal substrates are commonly utilized in high-performance electronic devices such as high-frequency circuit boards, power electronics, and applications requiring high temperatures. The copper substrates were utilized in their as-received state and were subsequently cut to the required size. The dimensions of the substrate are 30 mm (l), 30 mm (w), and 0.05 mm (t) as shown in Fig.1. In order to prevent the copper substrates from oxidizing and promoting adhesion between the coating and substrate, it was etched with sandpaper as illustrated in Fig. 2.

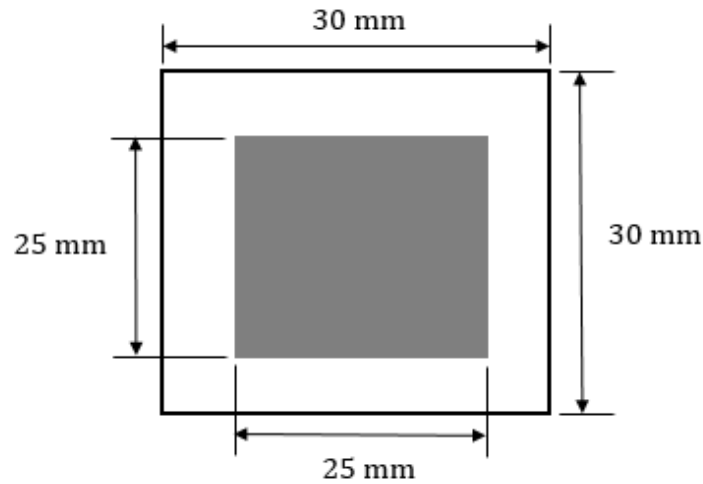


Fig. 1 Dimension of the copper substrate

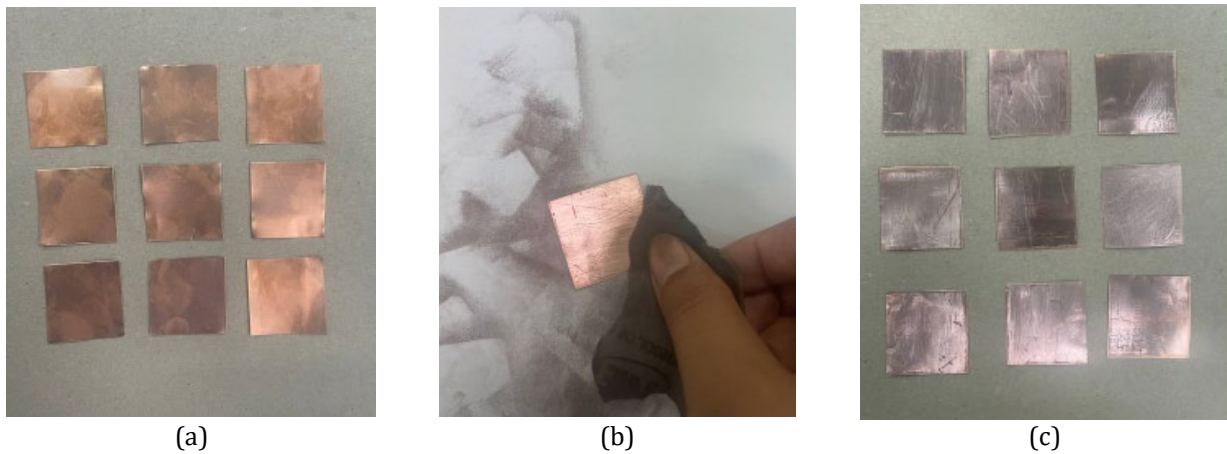


Fig. 2 Preparation of copper substrate; (a) Copper substrate; (b) Copper substrate etched with sandpaper; (c) Copper substrate after etched with sandpaper

2.3 Printing Method Process

The hybrid paste was printed on copper substrates as shown in Fig. 3. The hybrid GNP paste was placed on the selected grid (20mm x 20mm) and applied using a squeegee. The paste was printed on the copper substrate. After the printing was completed, the process was repeated on each sample.

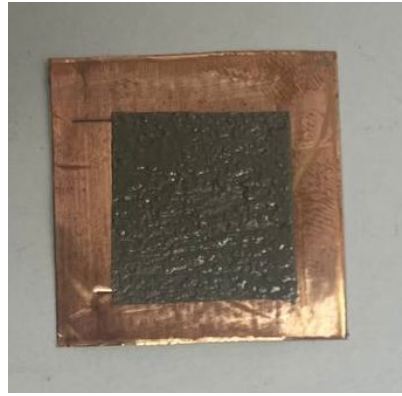


Fig. 3 GNP hybrid paste printed on copper substrate

2.4 Curing Process

The curing procedure is a post-treatment that is performed after the preparation of the hybrid conductive ink sample. The curing step is essential because it helps strengthen the bond that exists between the filler particles and the binder. For best conductivity, graphene must undergo a curing procedure to reduce porosity between particles and form a seamless conductive track [28].

The primary goal of the curing time experiment is to determine the optimal curing time and temperature for filler loading and observe the impact of various curing durations on the hybrid conductive ink. The time for curing was done from four (4) to six (6) hours with each sample interval of 1 hour. This time span allows a thorough investigation of the impact of curing time on the characteristics of the hybrid conductive ink, including any possible changes that might take place during this period. In addition, initiating the observation period at four (4) hours establishes a standard for comparison, while extending the length to six (6) hours allows for the detection of any additional alterations or patterns that may arise with longer periods of curing. The hybrid conductive ink was cured at temperatures of 240°C, 250°C and 260°C. 250°C of curing temperature was set as a baseline based on research done by [28]. In this research, the temperature was adjusted to 240°C, which is 10°C lower than the previous setting, and 260°C, which is 10°C higher than the prior setting of the baseline. The purpose of this adjustment was to investigate the influence of temperature fluctuations on the curing times of hybrid conductive ink. The samples underwent testing to assess their thermal, electrical, and mechanical properties. An experimental study was carried out to determine the optimal duration of curing for the GNP hybrid formulation in terms of resistivity and adhesion ability.

The prevalent approach for the curing process is heating in an oven. The sample was placed in an oven at a specific temperature and time to prevent any adverse effects on the formed substance and substrates. The parameters for samples for the adhesion test are shown in Table 3. After curing, all samples were normalized at room temperature for approximately 24 hours. The resistivity of the conductive ink was determined when the process was completed. Normalizing is the procedure of heating a material above a certain temperature and subsequently cooling it in the open air. The goal of normalizing the heat temperature is to enhance the mechanical characteristics of the material by optimizing the microstructure [28].

Table 3 Parameter of the adhesion test sample

Sample	Temperature (°C)	Curing duration
Sample 1	240	4 hours
Sample 2	240	5 hours
Sample 3	240	6 hours
Sample 4	250	4 hours
Sample 5	250	5 hours
Sample 6	250	6 hours
Sample 7	260	4 hours
Sample 8	260	5 hours
Sample 9	260	6 hours

2.5 Crosshatch Adhesion Test and Visual Inspection

Crosshatch Adhesion Test is a quantitative method used to measure the adhesion strength of coatings, while visual inspection provides a qualitative assessment by visually examining the coated surface for any signs of coating failure or defects. It was conducted following the guidelines of ASTM D6677 standard which specifies the examination of thin film adhesion using a knife equipped with multiple blades. The adhesion test, also known as the thin film peel test, is a commonly used technique to evaluate the adhesion strength of thin films because of its simplicity, effectiveness, and comprehensibility [29].

This method requires the use of a specific cutting tool to produce a grid-like pattern on the surface of the thin film. The cutting tool typically consists of multiple sharp blades arranged in a grid pattern. The blade tool used for this test has seven razor blades as shown in Fig. 4. The cutting tool was positioned at a 90° angle to the test panel's surface and consistent pressure was exerted to create a sequence of parallel cuts on the surface. Next, the cutting tool was realigned at a right angle to the initial cuts and made further parallel cuts to form a grid design. Then a scotch tape was applied over the crosshatched area, ensuring that it adhered firmly to the surface. The tape was pressed down to ensure good contact with the substrate. The tape was then peeled off rapidly and consistently, and the surface was examined for any signs of coating failure or detachment. The entire procedure is depicted in Fig. 5.

Adhesion performance is evaluated based on predetermined criteria using the ASTM 6677 standard. This standard uses an adhesion scale which ranges from 0B to 5B, where 5B represents the best adhesion. Table 1 displays the adhesion test rating and example. This method helps in assessing the adhesion quality and integrity of coatings or films on substrates.



Fig. 4 Blade tool with seven razors

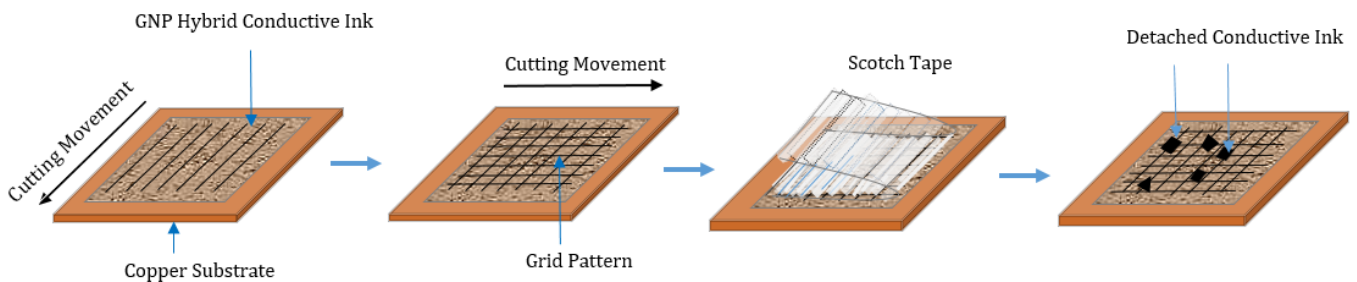
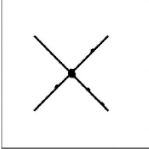
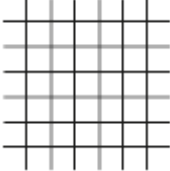
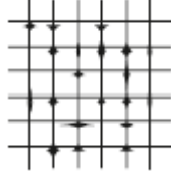
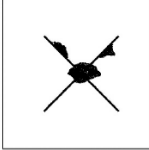
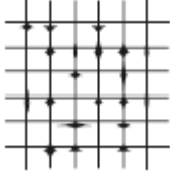
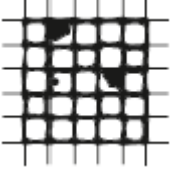
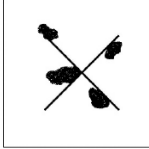
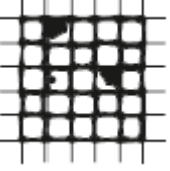
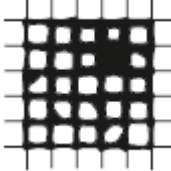
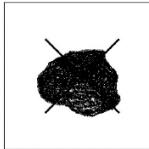
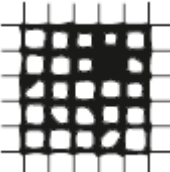
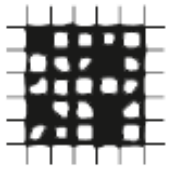
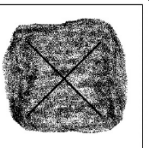

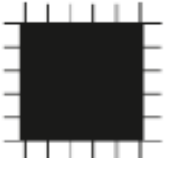


Fig. 5 Cross hatch adhesion test

Table 4 ASTM classification of adhesion test results

ASTM Classification	Description	Surface Of X-Cut Area	Remove %	Minimum	Maximum
5B	No peeling or removal		0		

4B	Trace peeling or removal along incisions or intersection		A cross-cut area <5%		
3B	Jagged removal along incisions		A cross-cut area 5% to 15%		
2B	Jagged removal along most of incisions		A cross-cut area 15% to 35%		
1B	Removal from most of the area of the X under the tape,		A cross-cut area 35% to 65%		
0B	Removal beyond the area of the X.		A cross-cut area >65%		

2.6 Curing Time and Temperature in Electrical Properties

The curing process for the hybrid conductive ink was conducted at various temperatures and durations. Specifically, the hybrid conductive ink paste was cured at 240°C, 250°C, and 260°C for four (4), five (5), and six (6) hours, respectively, at each temperature setting. Varying different curing temperatures and times in this research is to determine the most appropriate curing temperature and time and to investigate the impact of various curing periods on the adhesion properties of the hybrid conductive ink.

In this research, the curing time was set to range from four (4) to five (5) hours, with each sample interval of one (1) hour, so that each constant change in time would be able to show how the qualities of the material depend on time [30, 31]. The resistance measurements were taken using a Two-Point Probe at each point as shown in Fig. 6. This procedure was repeated for all samples in this research.

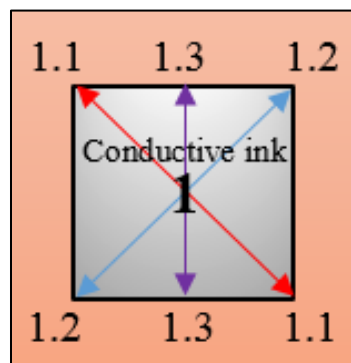


Fig. 6 Three points of printed GNP hybrid on copper substrates measurement taken using Two-Point Probe

3. Results and Discussion

3.1 Curing Temperature and Time

In this research, 250°C curing temperature was set as the baseline. The resistivity of the ink at the baseline curing temperature of 250°C is expected to reflect the typical electrical characteristics observed under standard curing conditions. This serves as a reference point for comparison with other temperature and duration variations. Table 5 shows the average resistance and average resistivity measurement data obtained at a temperature of 250°C for curing durations of four (4), five (5), and six (6) hours that were set as the baseline or as a reference point for comparison with other temperature and duration variations. The data is graphically depicted in Figure 7. The graph illustrates the outcomes of the curing duration for samples subjected to a temperature of 250°C. This research utilized a curing temperature of 250°C as the baseline and various curing durations were examined at this specific temperature. The curing parameter had been proven to affect adhesion [32, 33].

The findings obtained at a temperature of 250°C demonstrate a significant correlation between resistivity and the duration of curing. The observed pattern indicates an incremental enhancement in the ink's conductivity as the length of curing increases. The resistivity was measured at $3.0 \times 10^{-5} \Omega.m$ after four (4) hours of curing, while the resistivity dropped to $2.4 \times 10^{-5} \Omega.m$ after five (5) and six (6) hours of curing. The adhesion improves further at elevated curing duration [21]. Increasing the period of curing allows for a longer period of time for the ink to undergo complete cross-linking and polymerization processes. This leads to the formation of stronger intermolecular connections between the ink and the surfaces of the substrate. Based on research by Merilampi [21], the solvents efficiently evaporate starting from curing duration at 120°C and above, causing the silver particles to approach one other and create more conductive pathways for the movement of electrons between particles.

This is caused by the prolonged duration of curing potentially facilitating the elimination of remaining solvents or impurities, thereby increasing the conductivity of the ink. The stability of the conductive films over an extended period of time was influenced by the temperature at which they were cured [34]. The extended duration of curing enhances the adhesion between the conductive filler particles, leading to a more effective conducting network within the ink. Prolonged curing strengthens the bond between the filler particles and the surrounding matrix by giving the matrix additional time to fully cross-link and solidify [41].

In order to show how the qualities of the material depend on time, the data of resistivity of the samples in this study were fixed at curing duration, but varying the curing temperature between 240°C, 250°C, and 260°C. The curing durations in this research were set at four (4), five (5), and six (6) hours. This experimental setup allows the assessment of how temperature changes affect the resistivity of the conductive ink.

Table 6 displays the average resistance and average resistivity measurements acquired after curing for four (4) hours at temperatures of 240°C, 250°C, and 260°C, respectively. The data presented in the graph as shown in Fig. 8 provides insights into the influence of curing temperature on the resistance of the sample. The data illustrates the fluctuations in resistance and resistivity. This is because the thermal characteristics are significantly improved after high-temperature curing [36]. Resistivity exhibits a substantial drop as the curing temperature increases from 240°C to 260°C. More precisely, when the temperature reaches 240°C, the electrical resistivity measures $4.2 \times 10^{-5} \Omega.m$, and when it increases to 260°C, the resistivity decreases to $2.4 \times 10^{-5} \Omega.m$. It reveals a clear inverse relationship between curing temperature and resistivity, with higher temperatures corresponding to lower resistance values.

As mentioned earlier, the thermal properties are greatly enhanced during high-temperature curing as a result of the improved bonding and restructuring of the conductive filler particles inside the ink matrix. At higher temperatures, the curing process allows enhanced incorporation of the filler elements, leading to a stronger and more linked network. The increased structure enables effective heat movement and dissipation inside the material, resulting in improved thermal conductivity. The resistance exhibited a swift alteration following the elevation of the curing temperature. However, when the temperature was maintained at the same level, the change in resistance was comparatively less [32]. The finding aligns with the expected behaviour of materials that experience thermal processing, as greater temperatures enhance both bonding and conductivity [35].

Table 5 Data of resistance and resistivity at curing temperature 250 °C

Curing Duration for Sample with Temperature 250°C	Average Resistance Ω	Average Resistivity $\Omega.m$
4 hours	0.5	3.0×10^{-5}
5 hours	0.4	2.4×10^{-5}
6 hours	0.4	2.4×10^{-5}

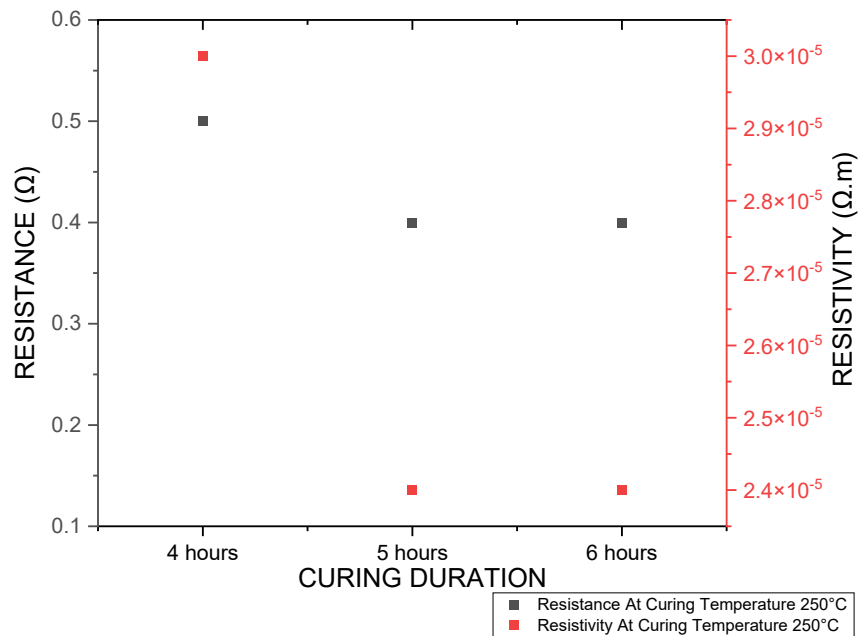


Fig. 7 Resistance and resistivity at curing temperature 250°C

Table 6 Data of resistance and resistivity at curing duration of 4 hours

Curing Temperature for Sample at 4 hours	Average Resistance Ω	Average Resistivity $\Omega.m$
240°C	0.7	4.2×10^{-5}
250°C	0.5	3.0×10^{-5}
260°C	0.4	2.4×10^{-5}

Tables 7 and 8 present the average values for resistance and resistivity measurements obtained after curing for 5 and 6 hours at temperatures of 240°C, 250°C, and 260°C, respectively. Both outcomes demonstrate the same pattern. The given data in Table 7 demonstrates the resistivity of the conductive ink at various curing temperatures at a fixed duration of 5 hours. The resistivity was measured at $3.6 \times 10^{-5} \Omega.m$ at a temperature of 240°C, and it reduced to $2.4 \times 10^{-5} \Omega.m$ at a temperature of 250°C. Remarkably, as the temperature reached 260°C, the resistivity experienced a significant increase to $8.4 \times 10^{-5} \Omega.m$. The data for the 6 hours curing duration exhibits a similar trend to the 5 hours curing duration. The resistivity of the material was measured to be $3.6 \times 10^{-5} \Omega.m$ at a temperature of 240°C, and it decreased to $2.4 \times 10^{-5} \Omega.m$ when the temperature was increased to 250°C. Nevertheless, as the temperature reached 260°C, the resistivity exhibited a noticeable rise to $6.6 \times 10^{-5} \Omega.m$. This implies the existence of an ideal temperature for the curing process that results in the lowest resistivity, and in this particular example, a temperature of 250°C is the most efficient. The increase in resistivity at 260°C may be related to possible deterioration caused by prolonged exposure to such temperature. This involves procedures like heat deterioration, which may change the molecular composition of the substance or the formation of defects and impurities that hinder the conductivity of electricity across the specimen. Moreover, prolonged exposure to high temperatures can cause alterations in the conductivity processes of the material or the formation of obstacles to the transit of electrons, resulting in an increase in resistance and resistivity. Higher temperatures may necessitate shorter times to achieve better efficiency, but longer curing time increases the potential for material deterioration [38].

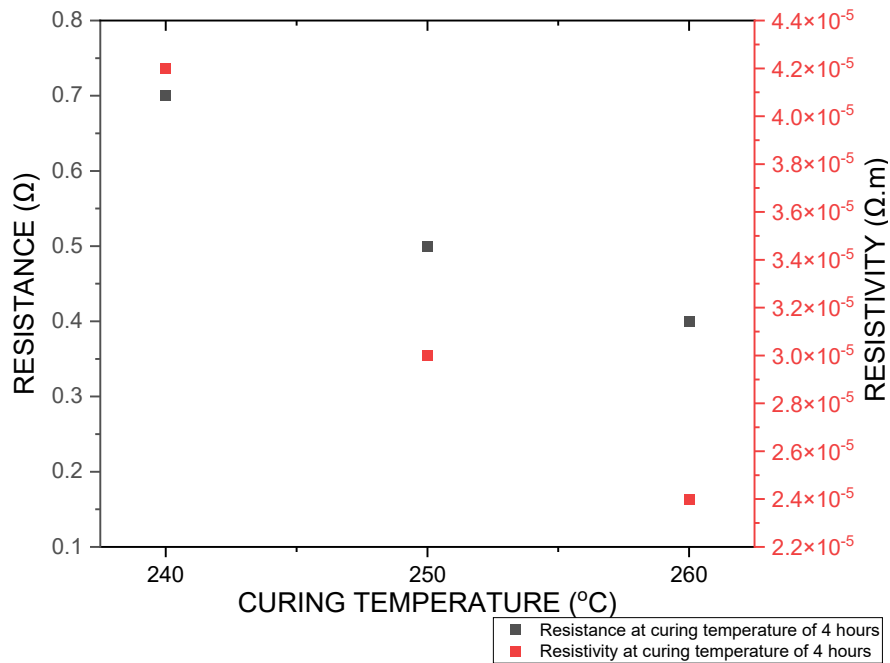


Fig. 8 Resistance and resistivity at curing duration of 4 hours

Table 7 Data of resistance and resistivity at curing duration of 5 hours

Curing Temperature for Sample at 5 hours	Average Resistance Ω	Average Resistivity Ω.m
240°C	0.6	3.6×10^{-5}
250°C	0.4	2.4×10^{-5}
260°C	1.4	8.4×10^{-5}

Table 8 Data of resistance and resistivity at curing duration of 6 hours

Curing Temperature for Sample at 6 hours	Average Resistance Ω	Average Resistivity Ω.m
240°C	0.6	3.6×10^{-5}
250°C	0.4	2.4×10^{-5}
260°C	1.1	6.6×10^{-5}

The graphs in Fig. 9 and Fig. 10 demonstrate notable fluctuations in the resistivity of the GNP hybrid conductive ink when subjected to various curing temperatures for a consistent duration of 5 and 6 hours. Significantly, the resistivity demonstrates a clear and distinct pattern throughout the temperature range that has been studied. Both results give a similar value when the temperature reaches 240°C, the resistivity is determined to be $3.6 \times 10^{-5} \Omega.m$. However, when the curing temperature is raised to 250°C, there is a clear reduction in resistivity to $2.4 \times 10^{-5} \Omega.m$, indicating enhanced electrical conductivity. Higher temperature promotes better bonding between the components of the conductive ink, including the graphene nanoparticles and silver flake. The strengthened bonding results in a more effective network of conductive pathways inside the ink, enabling better electron transport and thus lowering resistance. A conductive filler distribution network can provide a successful pathway for electron conduction [39]. The fluctuation mentioned earlier is illustrated in the graph, when the temperature reaches 260°C, the resistivity experiences a substantial increase, which diverges from the anticipated pattern as previously described.

The significant rise in resistivity at 260°C at five (5) and six (6) hours indicates that extended exposure to elevated temperatures could have detrimental consequences on the electrical characteristics of the ink, possibly as a result of thermal deterioration as discussed based on data at curing duration of 5 and 6 hours. As the temperature increases, the ions vibrate more intensely and more frequently as shown in Fig 11. The oscillating atoms and electrons clash, resulting in difficulties for the electrons to flow. The findings indicate that the resistance of the conductive ink shows significant variation in response to changes in curing temperature. The resistivity decreases as the curing temperature rises from 240°C to 250°C, indicating that higher temperatures

enhance the conductivity of the ink. Nevertheless, as the temperature reaches 260°C, the resistivity begins to increase anew. This suggests that if the material is exposed to even higher temperatures for a long period of time, its electrical performance may deteriorate. Therefore, the ideal temperature for getting the lowest resistivity seems to be around 250°C.

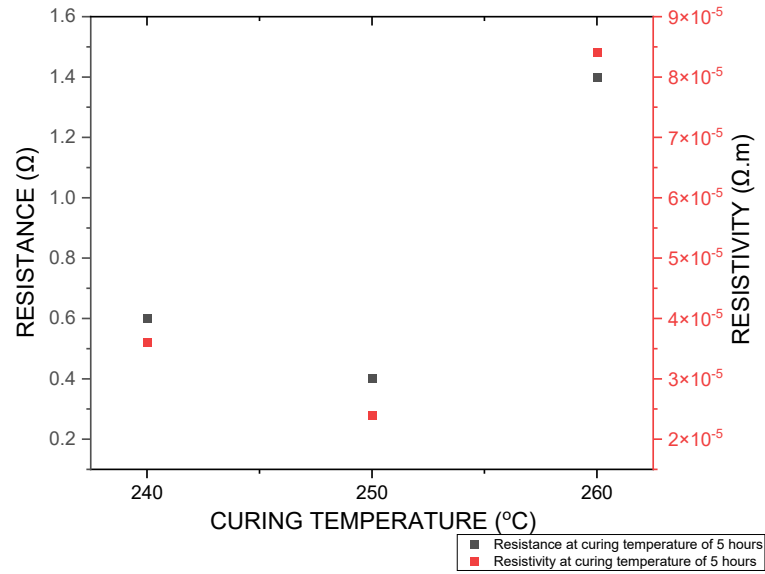


Fig. 9 Resistance and resistivity at curing duration of 5 hours

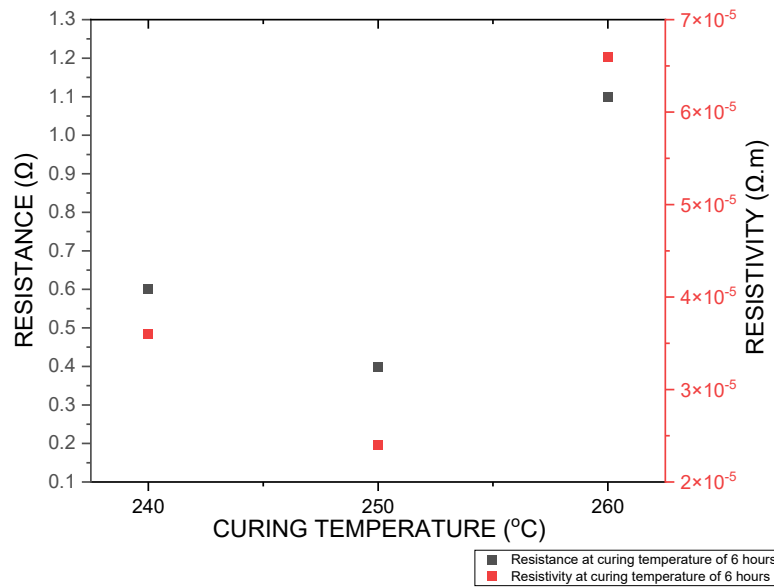


Fig. 10 Resistance and resistivity at curing duration of 6 hours

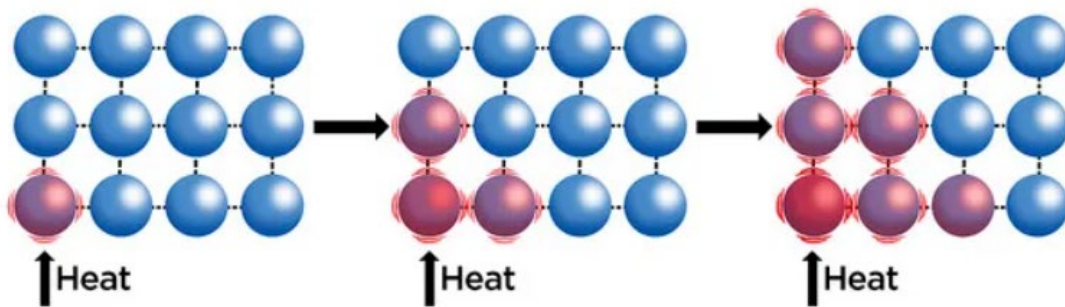






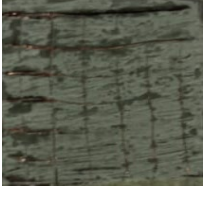




Fig. 11 Vibrating atoms when temperature increases [41]

3.2 Crosshatch Adhesion Peel Test

Visual inspection enhances the Crosshatch Adhesion Peel Test by distinguishing various failure causes and types of coating deterioration. Common failure modes consist of cohesive failure, adhesive failure, and other flaws like blistering, cracking, or delamination. The failure modes offer qualitative insights into the adhesion quality and integrity of the coating. The crosshatch adhesion test was performed using GNP hybrid conductive ink that was cured at a temperature of 240°C, 250°C, and 260°C for durations of 4, 5, and 6 hours, respectively. The results, in accordance with the ASTM 6677 standard as shown in Table 9, showed variable adhesion strengths based on the length of time the curing process occurred.

Table 9 Crosshatch Adhesion Peel Test result at 240°C

Curing Time	Adhesion Test Result at 240°C	ASTM Result at 240°C	Adhesion Test Result at 250°C	ASTM Result at 250°C	Adhesion Test Result at 260°C	ASTM Result at 260°C
4 hours		4B		3B		4B
5 hours		3B		4B		3B
6 hours		3B		2B		2B

As depicted in Table 9, at 240°C after 4 hours of curing, the adhesion strength was evaluated as 4B, which suggests a slight peeling or removal along the incisions or intersections. By increasing the curing time to 5 hours, the adhesion strength marginally dropped to 3B, indicating rough removal along incisions with a cross-cut area ranging from 5% to 15%. Interestingly, maintaining the curing duration at 6 hours resulted in a similar adhesion strength of 3B, suggesting constant adhesion performance despite the extended curing time. These data indicate that the period of curing affects the strength of adhesion, especially during the transition from 4B to 3B between 4 and 5 hours. However, prolonging the curing time beyond 5 hours is unlikely to significantly change the adhesion properties of the GNP hybrid conductive ink. Higher curing temperatures facilitated the dissolution [37]. Dissolution is the phenomenon when a solid substance breaks down into a liquid, creating a uniform mixture in which the solid particles disperse uniformly throughout the liquid phase. Such behavior aligns with the fundamental principles of materials science, wherein elevated temperatures promote enhanced bonding and conductivity.

The results at a temperature of 250°C demonstrated varied levels of adhesion strengths associated with varied curing times. After 4 hours of curing, the adhesion strength was evaluated as 3B, which indicates the removal along incisions was rough and jagged, covering an area of 5% to 15%. By increasing the curing time to 5 hours, the adhesion strength improved to 4B, indicating the occurrence of slight peeling or removal at incisions or junctions with a cross-cut area of less than 5%. However, when the curing period was increased to 6 hours, there was a significant reduction in adhesion strength to 2B. This suggests that the removal along most of the incisions was uneven, resulting in a cross-cut area ranging from 15% to 35%. The results emphasize the substantial impact

of the curing duration on the adhesion capabilities of the conductive ink. The drop in adhesion strength found at the 6-hour mark can be caused by excessive drying or curing, which results in diminished bonding between the ink and substrate. Extended curing durations and overly elevated curing temperatures will result in the brittleness of the conductive ink

The result at a temperature of 260°C showed significant variation in adhesion strength based on different curing times. After 4 hours of curing, the adhesion strength was evaluated as 4B, which suggests slight peeling or removal at incisions or junctions with a cross-cut area of less than 5%. With an increase in the curing duration to 5 hours, the adhesion strength showed a modest decrease to 3B. This indicates that there was jagged removal along incisions with a cross-cut area ranging from 5% to 15%. Significantly, increasing the curing period to 6 hours resulted in a significant decrease in adhesion strength, with a rating of 2B. This rating indicates that there was rough removal throughout most of the incisions, with a cross-cut area ranging from 15% to 35%.

These findings highlight the substantial influence of the length of curing on the adhesion performance of the conductive ink. The decrease in adhesion strength observed at the six (6) hours at curing temperatures of 250°C and 260°C, which is caused by excessive curing that leads to heightened brittleness or diminished bonding between the ink and substrate. Elevated temperatures can lead to the deterioration or oxidation of graphene, causing an adverse impact on its electrical conductivity and an increase in resistance. Conductive ink's resistance decreases at curing temperatures above 150°C but the conductive ink becomes brittle when curing times are prolonged or the temperature is raised too high [41]. The brittleness is caused by extended curing periods and high temperatures resulting in an excessive amount of solvent evaporating, which makes the ink harder and less flexible. High temperatures can also hasten the chemical reactions that occur within the ink matrix, resulting in the formation of cross-linking connections that contribute to the material's stiffness. These factors contribute to the brittleness of the conductive ink, making it more prone to cracking or delamination.

4. Conclusion

The results of the studies demonstrate that increased curing temperatures have an adverse effect on the adhesive strength of the conductive ink, highlighting the correlation between curing conditions and adhesion qualities. The findings indicate that the resistivity of the conductive ink varies significantly with changes in curing temperature. The primary aim of varying the curing temperature and experiment time is to determine the optimal curing temperature and time and investigate the impact of various curing periods on the adhesion properties of the GNP hybrid conductive ink. Specifically, the resistivity decreases as the curing temperature increases from 240°C to 250°C, suggesting that higher temperatures promote better conductivity of the ink. However, at 260°C, the resistivity increases again, indicating that prolonged exposure to even higher temperatures may lead to a deterioration in electrical performance resulting in the optimal curing temperature for achieving the lowest resistivity appears to be around 250°C.

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Conflict of Interest

Authors declare that there is no conflict of interest regarding the publication of the paper.

Author Contribution

*The authors confirm their contribution to the paper as follows: **study conception and design:** Norida Mohammad Noor, Mohd Azli Salim; **data collection:** Nor Azmmi Masripan; **analysis and interpretation of results:** Norida Mohammad Noor, Adzni Md. Saad; **draft manuscript preparation:** Norida Mohammad Noor, Mohd Azli Salim, Chonlatee Photong, Mohd Zaid Akop. All authors reviewed the results and approved the final version of the manuscript.*

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