Journal of Advanced Research in Applied Mechanics 119, Issue 1 (2024) 1-12

Journal of Advanced Research in Applied Mechanics

Journal homepage:
https://semarakilmu.com.my/journals/index.php/appl_mech/index
ISSN: 2289-7895

Effect of GNP/Ag Stretchable Conductive Ink on Electrical Conductivity

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ARTICLE INFO

Article history:
Received 7 March 2024
Received in revised form 2 May 2024
Accepted 16 May 2024
Available online 30 June 2024

Keywords:
Stretchable conductive ink; graphene nanoplatelet; silver flakes; resistivity

ABSTRACT

This research aims to develop and formulate a highly thermal graphene hybridization conductive ink combining graphene nanoparticles (GNP), silver flakes (Ag), and silver acetate (SA) as conductive fillers mixed with chemical and organic solvents. With improved properties, it overcomes the limitations of traditional materials while preserving their beneficial characteristics. The study evaluates how the resistivity and properties of the material change in response to environmental factors such as temperature and humidity and how these changes impact its performance in various applications. To develop a highly thermal graphene hybridization conductive ink, a new formulation of conductive ink was formulated using graphene nanoparticles (GNP), silver flakes (Ag), and silver acetate (SA) as conductive fillers mixed with organic solvents. In order to turn the batch of substances into a powder, they were sonicated and followed by stirring to form the mixture into a powder. Before curing at 250oC 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. Using a mesh stencil, the GNP hybrid paste was printed on copper substrates. With a scraper, the hybrid GNP paste was applied to the selected grid (3mm x 3mm) on three selected points of the substrate strip. In order to evaluate the performance, the resistivity of the hybrid GNP conductive ink at room temperature was set as the baseline and compared to the resistivity readings obtained at varying temperatures-humidity levels. GNP hybrid room temperature baseline and GNP hybrid after applying different temperature-humidity were compared in terms of electrical and mechanical properties. The average resistivity measurement at all points of the sample remained stable or decreased as the temperature increased. It demonstrates that the electrical conductivity of the ink degrades significantly as the temperature-humidity increases. This indicates that the ink is able to maintain its structural integrity and properties within certain temperature ranges. This signifies that a hybrid conductive ink has good thermal stability. Future work should investigate the strategies for improving the ink's performance under mechanical deformation, such as the use of additives or novel printing techniques.

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https://doi.org/10.37934/aram.119.1.112

1. Introduction

Printed electronics is one of the fastest-growing technologies today, with applications in healthcare, aerospace, automotive industries, building and construction, energy and photovoltaic, textiles, consumer electronics, and smart packaging and logistics as illustrated in Figure 1 [1]. However, the development of high-performance stretchable conductive inks is critical to achieving the necessary flexibility and conductivity for these applications. Graphene nanoparticles (GNP) and silver flake (Ag) particles are commonly used in stretchable conductive inks due to their unique electrical properties. This study aims to investigate the effect of GNP/Ag concentration on the electrical conductivity of stretchable conductive inks. Specifically, it focuses on GNP/Ag-based inks and explored how the concentration of these particles affects the ink's electrical conductivity. By understanding the impact of GNP/Ag concentration on the electrical conductivity of these inks, more effective formulations for stretchable conductive inks can be developed to meet the demands of flexible and wearable electronics.

Fig. 1. Application areas of flexible and printed electronics [1]

Conductive inks are based on complicated, multi-component compositions. In the production of conductive ink, the conductive substance is the most crucial component. The selection of conductive materials is based on the physical properties of the to-be-printed pattern, such as adherence to the substrate, and the desired physiochemical properties of the inks, such as compatibility with the printing procedure. This includes nanoparticle printing limits on nozzle size, aggregation, stability, rheology, and electrical and mechanical properties [2-7]. Above all, the qualities and behaviour of inks are determined by their rheology and surface energy. The rheology of the ink must correspond to the planned printing technique, the precise environmental conditions, the printing equipment, and the production speed and parameters. Figure 2 provides a summary of the required characteristics of conductive inks.

Stretchable conductive inks have attracted increasing attention due to their potential applications in flexible and wearable electronics. Graphene nanoplatelets (GNPs) and silver nanoparticles (Ag NPs) are among the most commonly used materials for fabricating stretchable conductive inks due to their excellent electrical conductivity and mechanical properties [6, 7].

Stretchable conductive inks are essential materials for the realization of flexible and wearable electronics. Over the years, several researchers have made significant efforts in developing and improving these inks' electrical and mechanical properties to achieve optimal performance. One popular approach for enhancing stretchability is to use a combination of conductive nanoparticles, such as silver, copper, and gold, with elastomers. Most researchers reported the development of a highly stretchable conductive fabric made of silver nanoparticles embedded in polyurethane.

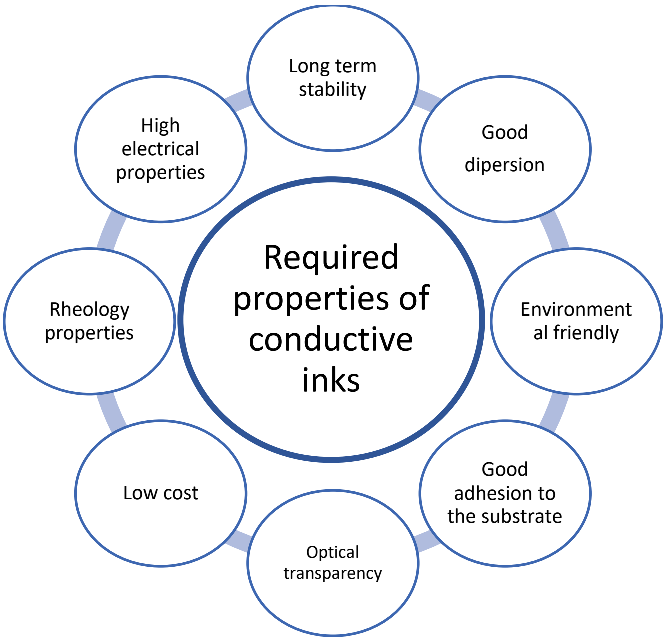


Fig. 2. Summary of required properties of conductive inks [19]

Other researchers have explored the use of carbon-based nanoparticles, such as graphene and carbon nanotubes, in stretchable conductive inks. Some articles provided a review of the progress in developing stretchable conductive inks using these materials, including recent advancements in ink formulations and applications [8-15].

Stretchable conductive inks are typically manufactured using liquid-phase printing techniques such as gravure, flexographic, screen printing, and inkjet printing. The inkjet printing technique is gaining popularity due to its simplicity and cost-effectiveness in printing several electrical and optoelectronic devices on a variety of rigid and flexible substrates. The greatest benefit of this technology is that it does not necessitate costly and time-consuming processes such as plating, masking, and etching, which are often employed in conventional methods such as photolithography [16-19].

The increase in the number of articles published on flexible and stretchable electronics over the last decade also demonstrates the promising future of printed electronics, especially when taking into account their mechanical durability, improved foldability, and portability. In recent years, there has been an increasing interest in researchers in developing different fabrication techniques for mass-producing high-quality electronic devices using the roll-to-roll (R2R) method processing on low-cost and flexible substrates [20]. Printed flexible organic and electronics are expected to grow in popularity in the future due to the fact that flexible substrates allow for printed electronics with greater mechanical robustness [4].

In the context of stretchable conductive inks, electrical conductivity and resistivity are essential properties that determine their performance in various applications. Researchers often study the effect of different factors, such as temperature, pressure, and strain, on these properties to optimize the ink's performance. Environmental factors like temperature and humidity have an impact on the electrical conductivity and resistivity of stretchable conductive inks. Temperature is a critical environmental factor that can affect the performance of stretchable conductive inks. As the temperature changes, the resistance of the conductive ink can increase or decrease, depending on the type of ink and the specific temperature range. Saleh *et al.,*[21] investigated the temperature-dependent electrical conductivity of a stretchable silver-nanoparticle-based ink and found that the electrical conductivity increased with temperature up to a certain point, after which it decreased due to particle agglomeration.

Humidity is another important environmental factor that can affect the electrical conductivity and resistivity of conductive inks. High levels of humidity can lead to water absorption by the ink, which can alter its electrical properties. Saleh *et al.,* [21] also studied the effect of humidity on the electrical conductivity of graphene-based stretchable conductive inks and found that the conductivity decreased significantly as the humidity level increased.

It is important to consider environmental factors like temperature and humidity when designing and testing stretchable conductive inks for practical applications. These factors can significantly impact the performance and reliability of the ink, and understanding their effects can help optimize the ink's performance and ensure that it functions well under a range of environmental conditions.

In summary, GNP/Ag stretchable conductive inks have shown promising electrical conductivity and mechanical properties for flexible and wearable electronics. Understanding the electrical conductivity and resistivity of stretchable conductive inks is essential for developing robust and reliable electronic devices that can withstand mechanical deformation. Furthermore, the development of conductive inks with high electrical conductivity and low resistivity will enable the creation of more complex and high-performance electronic devices.

2. Methodology

Constituents used to prepare the highly thermal graphene hybridization conductive ink consisted of graphene nanoparticle (GNP), silver flake (Ag) and silver acetate (SA) as the main fillers, ethanol as the chemical solvent, and 1-Butanol, and terpineol as the organic solvents that act as a binder.

The parameters used to prepare highly thermal graphene hybridization conductive ink are tabulated in Table 1 and the composition in which the different constituents are 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. 1 set of compositions is defined as a combination of ingredients used to make a single portion of the conductive ink. In this research, 10 sets were used, which means 10 portions or 10 times the amount of specified ingredients in 1 set were combined to make the paste.

Table 1
Constituents of highly thermal graphene hybridization conductive ink

Type	Material
Filler material	GNP powder 5 µm nanoparticle size Silver flakes (Ag) Silver Acetate (SA)
Chemical Solvent	Ethanol 99.99%
Organic Solvent as binder	I-Butanol Terpineol

Table 2
Composition of different constituent's mixture according to the weight for 1 set

	GNP (g)	Silver Flake (g)	Silver Acetate (g)	Ethanol (ml)	I-Butanol (drop)	I-Tripeneol (drop)
1 set	0.05	0.4292	0.042	5	3	3

For the preparation of 10 sets, 0.5 g of GNP powder (xGnP H-5) with 5µm particle size was mixed with 50 ml ethanol in a beaker. During the course of the experiment, an aluminum foil cover was placed over the beaker to prevent the ethanol from evaporating. The GNP was sonicated in ethanol for 10 minutes. Then the mixture was added with 4.292 g of silver flake. The very fine texture of SA is weighted on PET and added to the GNP/ethanol mixture. It proceeded with the sonication for one (1) hour. During the sonication process, the dispersibility performance of GNP in ethanol and Ag was observed. The sample preparation of GNP/Ag Conductive ink is illustrated in Figure 2.

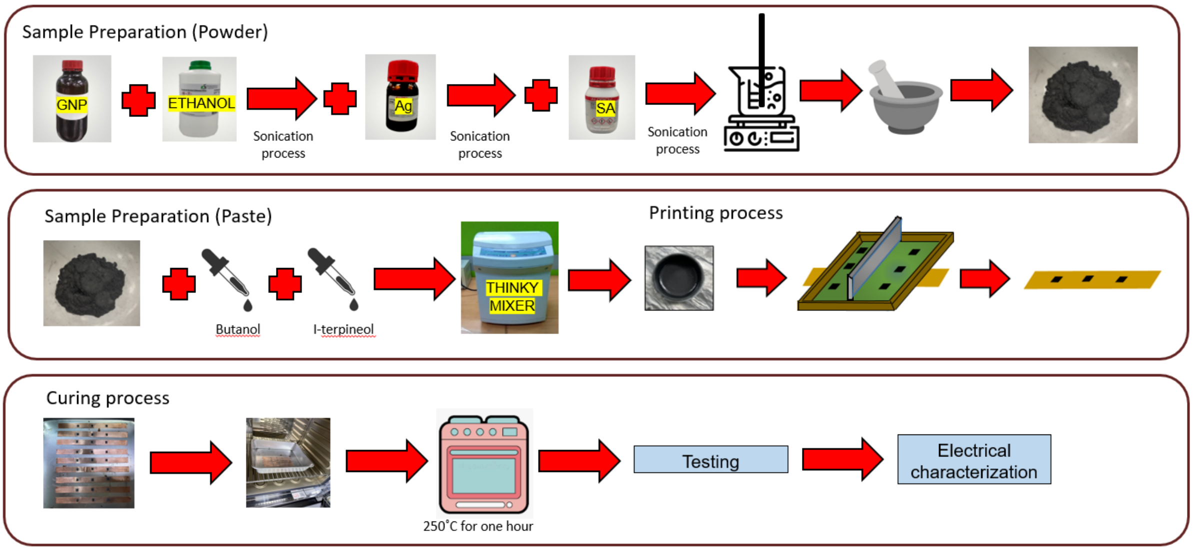


Fig. 2. Sample preparation of GNP/Ag Conductive ink

The mixture was then added with 0.42 g of silver acetate, SA, and sonicated for another hour. The solution was then heated on a hotplate at 70 °C while being stirred at 200 rpm until the excess ethanol evaporated. After being stirred, the mixture was put into a small white porcelain beaker and baked at 250 °C for one (1) hour to cure. After the curing process, the room-temperature mixture was pounded until it produced a fine powder. After that, the powder was put into a container so that it could be used in the preparation of the GNP hybrid paste.

For the preparation of the GNP hybrid paste, 45 drops of butanol and 45 drops of terpineol were added to the GNP hybrid powder of 4.68 g. The mixture of GNP hybrid powder with the organic solvent as a binder was weighted to ensure the right setting and then placed in the thinky mixer. Thinker mixer is a machine that demonstrates superior performance in dispersing nanoparticles in a variety of media while removing the air bubbles and produces thoroughly mixed compositions.

The GNP hybrid paste was then printed on copper substrates using a mesh stencil with a thickness of 10 µm. The hybrid GNP paste was then applied to a selected grid (3mm x 3mm) on three selected points of the substrate strip using a scraper as illustrated in Figure 3.

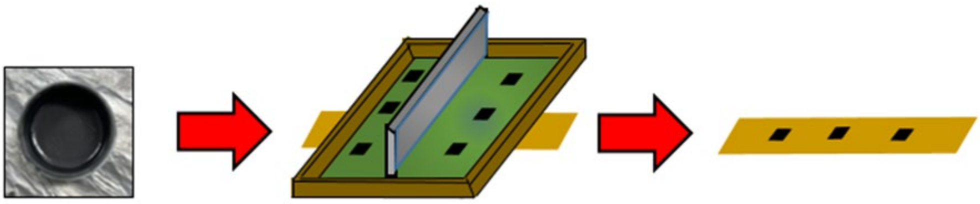


Fig. 3. Printing process using the mesh stencil

To test the performance of the GNP hybrid conductive ink, cyclic testing was conducted using a cyclic bending test and cyclic torsion test machines. The testing was performed in a heat chamber with different temperature-humidity settings based on the number of lamps. After each cycle (1000 cycle, 3000 cycle, and 5000 cycle), the resistance of the conductive ink was measured to determine the resistivity.

3. Results

The resistivity of a material is a measurement of its electrical resistance, and it can be affected by a variety of environmental factors such as temperature and humidity. To study these effects, a common approach is to measure the resistivity of a material at a reference or baseline condition. The resistivity of the hybrid GNP conductive ink at room temperature was set as the baseline and compared to the resistivity readings obtained at varying temperatures-humidity levels. Resistivity not only depends on the chemical composition of conductive ink but also on the shape of the printed ink, thickness, length, and cross-sectioned area [16].

The result of a GNP hybrid conductive ink at room temperature as a baseline refers to the measurement of the ink's electrical conductivity under standard and controlled conditions. The room temperature serves as a reference point for the electrical conductivity of the ink, as the temperature is a common and consistent condition that can be used to compare and evaluate the performance of the ink over time and under different conditions.

Based on Figure 4, the value of all samples has a slight difference at each point. With a low coefficient of variation (CV) the data has a small range of variability, which means that the data points are close to each other and the electrical conductivity of the ink is consistent across all the samples. It can be concluded that the electrical conductivity of the GNP hybrid conductive ink is stable and consistent under standard and controlled conditions. This means that the ink has a consistent electrical conductivity and does not change significantly even when multiple samples are tested under the same conditions.

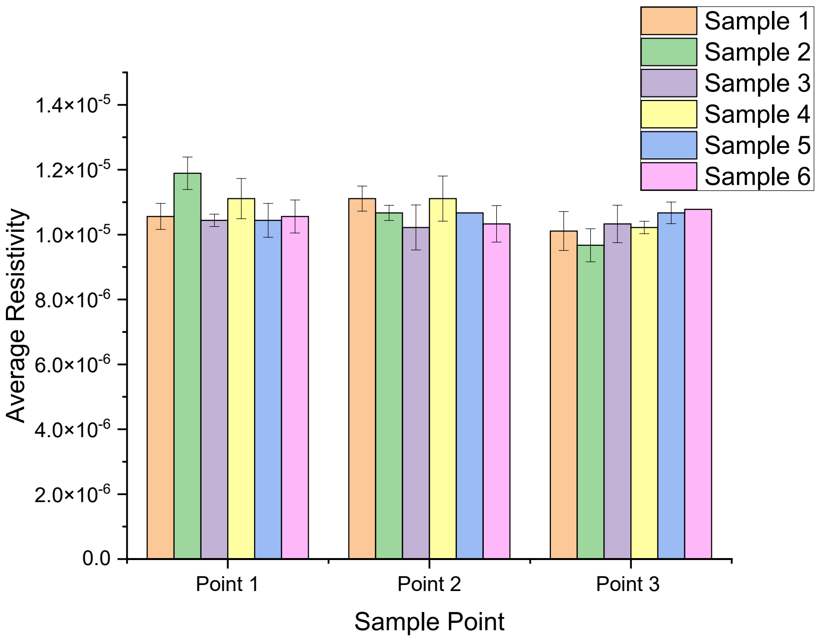


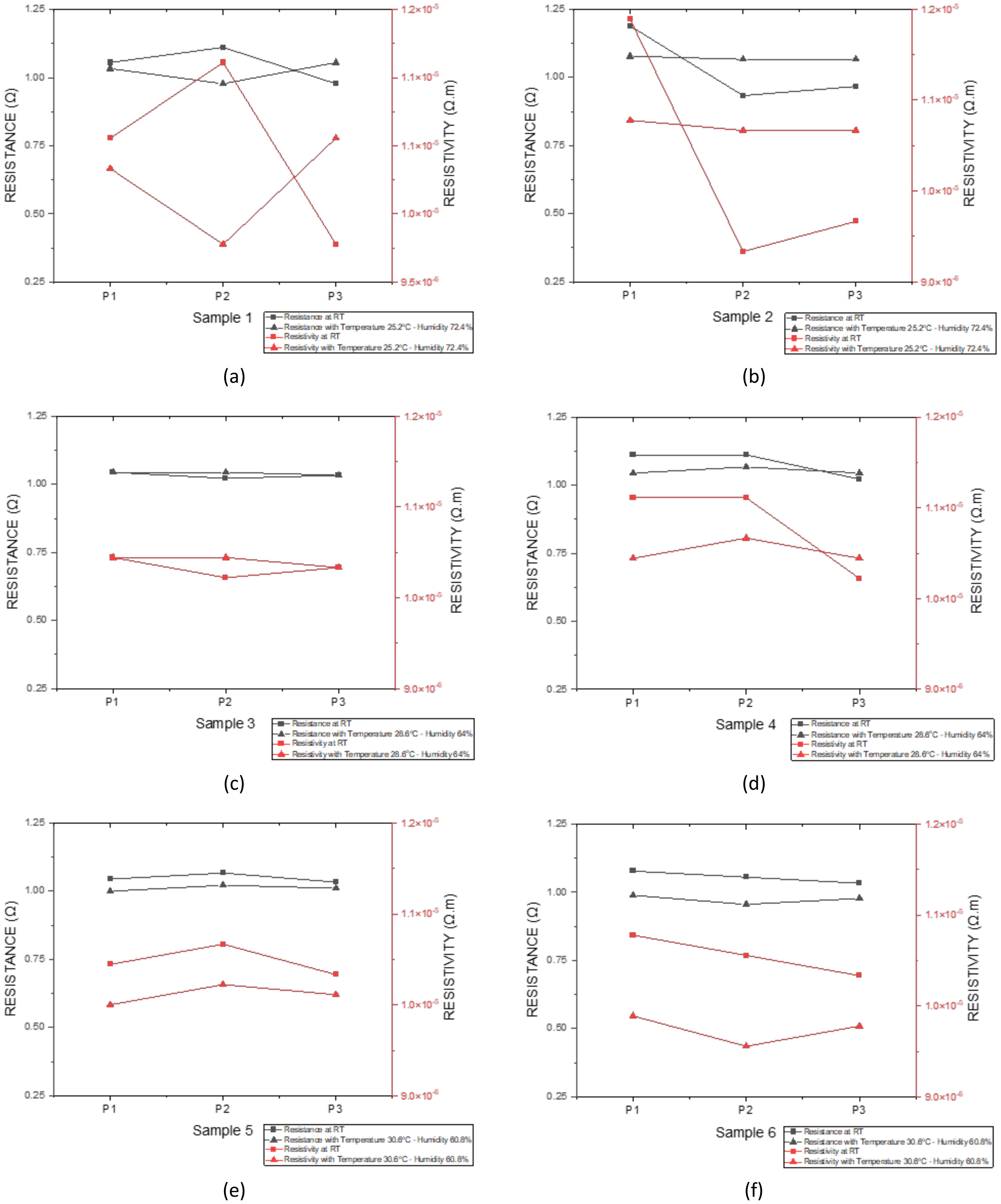
Fig. 4. GNP/Ag Room Temperature Baseline

Figure 5 shows the data of resistance and resistivity for all samples at room temperature and after different temperature-humidity levels were applied based on the number of lamps in the heat chamber. From the data, the average resistivity measurement at all points remains stable or decreases as the temperature increases except for sample 2 at point 3. The resistivity percentage change of increment is below 5% which shows no significant differences of resistivity value.

The resistivity value of the GNP hybridization conductive ink remains constant or decreases with the increasing temperature prior to cyclic testing. This finding is consistent with previous studies that reported a decrease in resistivity with the increasing temperature for conductive materials containing silver or graphene particles [11, 12].

The decrease in resistivity with the increasing temperature can be attributed to a decrease in the number of impurities or defects in the conductive ink at higher temperatures which leads to improved electron transport and reduced resistivity [10]. This phenomenon is often observed in metals and metal alloys, where increasing temperature can cause a reduction in the number of defects and an increase in the mobility of free electrons.

Figure 6 shows the increment of temperature and reduction of humidity during the cyclic bending testing, which has a significant effect on the resistance of conductive ink. Based on the average data of resistance and resistivity, as temperature-humidity increases, the resistance of the material slightly increases, except for the 6 lamps at cycle of 1000 with temperature of 30.6°C and humidity of 60.8%, which showed a significant increase in resistivity. It can be observed that at high temperature with low humidity can cause the ink to expand, which can lead to cracking and delamination. This causes a loss of electrical conductivity and increases resistance. Additionally, high temperatures cause the ink to degrade chemically, which also leads to an increase in resistance. These effects are more pronounced as the temperature increases. The failure connection between devices affects the electrical resistance and reduce the bonding strength between them when exposed to hot temperature and humid environment [7-9].



(a) (b)

(c) (d)

(e) (f)

Fig. 5. Measurement of Resistance and Resistivity based on Room Temperature and with different Temperature-Humidity

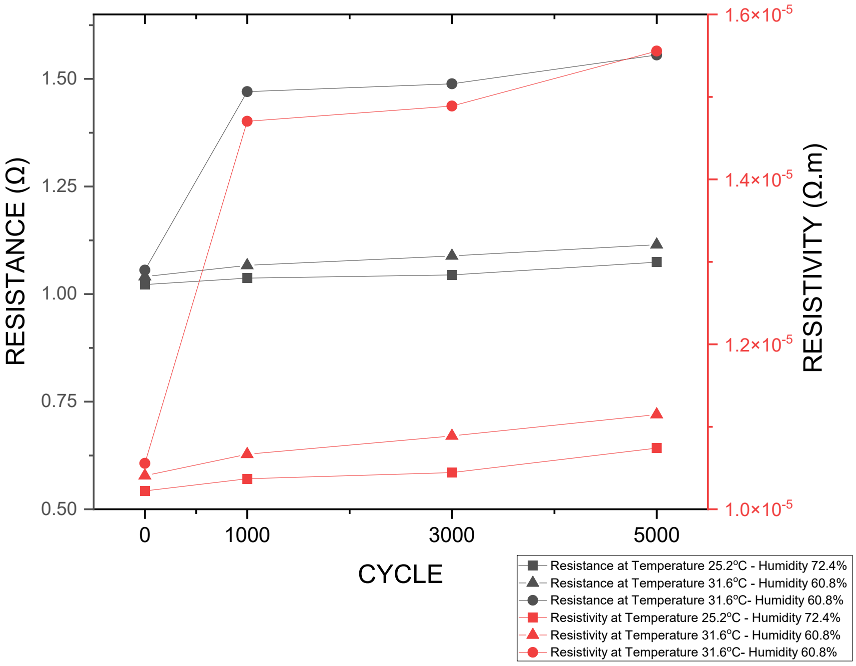


Fig. 6. Measurement of Resistance and Resistivity of Each Cycle Based On Bending Test with Different Temperature and Humidity

Figure 7 shows the value of average resistance and resistivity of samples after each cycle based on cyclic torsion test with different temperature-humidity. It shows an increasing value of resistance and resistivity with every progress of the torsion cycle, which displays a clear trend. In regards to temperature and humidity effects, the temperature-humidity differences between lamps 2, 4 and 6 were quite steady and the increase of resistance and resistivity under 6 lamps were quite significant. In cyclic torsion testing, due to the accumulation of plastic deformation and the increased likelihood of fatigue failure, the resistance of a material can increase as the number of cycles and temperature increase. As the specimens are subjected to repeated twisting, small defects or inhomogeneities in the material are amplified, these lead to the initiation and propagation of micro-cracks, which can cause a loss of strength and an increase in resistance. The significant increase in dislocation density and dispersion caused by the high imposed strain can be attributed to the resistivity increase after processing [19].

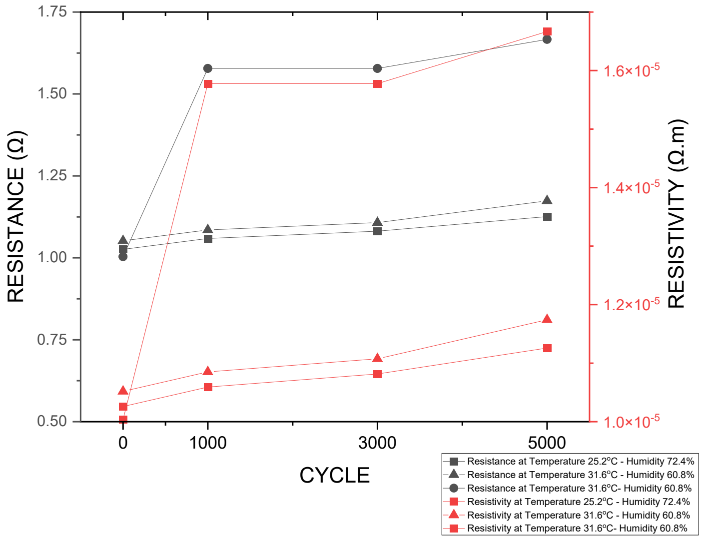


Fig. 7. Measurement of Resistance and Resistivity of Each Cycle Base on Torsion Test with Different Temperature and Humidity

The resistance and resistivity of the GNP hybridization conductive ink increase with each cycle of bending and torsion. This behavior is consistent with the results of previous studies that have been investigated related to the effect of mechanical deformation on the electrical properties of conductive inks [14, 16]. The increase in resistance and resistivity is due to the deformation of the ink's conductive pathways, which can lead to the formation of cracks or gaps in the ink layer. These defects can impede the flow of electrical current, resulting to an increase in resistance and resistivity [15].

These findings suggest that the GNP hybridization conductive ink is suitable for applications that involve cyclic bending or torsion, it is worth noting that the ink's performance can be improved through optimization of the ink formulation or printing parameters. For example, previous studies have reported that the use of a binder or the incorporation of a sacrificial layer can help to improve the mechanical durability of conductive inks [16].

The increase in resistance and resistivity of the GNP hybridization conductive ink with cyclic bending and torsion highlights the importance of considering the mechanical durability of conductive inks in flexible and wearable electronic applications. Future work should investigate the strategies for improving the mechanical durability of these inks, such as the use of additives or novel printing techniques.

4. Conclusions

Based on the correlation study of different temperature and humidity conditions on highly thermal GNP/Ag stretchable conductive ink, it was found that the ink's electrical resistivity decreased with the increasing temperature. The GNP hybridization conductive ink was considered thermally stable when its resistivity value remained constant or decreased with the increasing temperature before the cyclic testing. This indicates that the ink's electrical conductivity does not deteriorate significantly as temperature increases, indicating that the ink maintains its structural and electrical properties within a specific temperature range. This is a hallmark of a hybrid conductive ink with good thermal stability.

In conclusion, the finding of a constant or decreasing resistivity value of the GNP hybridization conductive ink with the increasing temperature prior to cyclic testing supports the use of these materials in high-temperature applications where low resistivity is critical. However, further research is needed to investigate the effect of temperature during cyclic loading to fully understand the performance of these materials in flexible and wearable electronic applications.

Acknowledgement

Thank you to Advanced Academia-Industry Collaboration Laboratory (AiCL) and Fakulti Kejuruteraan Mekanikal (FKM), Universiti Teknikal Malaysia Melaka (UTeM) for providing the laboratory facilities.

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