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Investigation of Functional Ageing of GNP/Ag Conductive Ink under Torsional Conditions

Muhammad Arieef Hussain[1], Mohd Azli Salim[2*], Nor Azmmi Masripan[2], Adzni Md. Saad[2], Mohd Zaid Akop[2], Chonlatee Photong[3]

[1] *Department of Mechanical Engineering, Politeknik Ungku Omar, MALAYSIA*
[2] *Fakulti Teknologi dan Kejuruteraan Mekanikal, Universiti Teknikal Malaysia Melaka, MALAYSIA*
[3] *Faculty of Engineering, Mahasarakham University, THAILAND*

*Corresponding Author: azli@utem.edu.my
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Graphene nanoplatelet (GNP), silver flakes (Ag), hybrid conductive ink, resistivity, cyclic torsion, ageing

Abstract

This study evaluated the ageing effects after enduring a cyclic torsional test on electrically high thermal Graphene Nanoplates (GNP)/Ag conductive ink. The Graphene Nanoplates (GNP) hybrid formulation contained silver flake (Ag) as an additional filler, 1-butanol and terpineol as organic solvents for the paste, and ethanol as a chemical solvent for the powder GNP hybrid synthesis. An in-house torsional cyclic test rig acted as an experimental apparatus to hold cyclic torsional tests in measuring the resistivity value of GNP/Ag conductive ink. The sample endured 1000, 2000, and 4000 cycles of repetitive cyclic torsion. The sample was left unattended in a controlled environment for ageing process observation in 15 weeks. Resistivity values for both tests were obtained by using a two-point probe multimeter. Data from the cyclic torsional test showed an increase in average resistivity as the cycles prolonged until 4000 cycles. The outcome of these experiments proved that Graphene Nanoplates (GNP)/Ag conductive ink manages to withstand material deformation and remain electrically stable after experiencing cyclic twisting and the ageing process. The recorded SEM images of the ink layer microstructure surface showed a direct correlation with the obtained sheet resistivity data. The samples that produced high sheet resistivity showed the presence of microcracks and defects on the ink layer The results of this study imply that repeated twisting in a thin film may lead to different mechanical and material modifications, thereby influencing its electrical and mechanical properties, which is vital to developing more reliable and stable stretchable electronics. Additional research on various twisting angles and ink compositions is recommended to have a more profound understanding of the characteristics and features of GNP/Ag conductive ink.

1. Introduction

Graphene Nanoplatelets (GNP) based supercapacitors are increasingly attractive in conjunction with the expected high surface area, high electron mobility, and other excellent characteristics of pure graphene [1]; [2]. Asiya et.al

[3], added that this material possesses superior physicochemical qualities, exceptional mechanical performance, and distinctive electrical and thermal conductivities. Graphene is a single layer of a 2D array of hybridized carbon atoms extracted from graphite layers, connected by sp2 bonds in a honeycomb pattern with a C-C bond length of 0.142 nm. GNP has the highest aspect ratio and specific surface area among all known nanomaterials. It may develop an uninterrupted electrical conductive network within the polymer matrix with minimal filler loading as a result of its specific surface area and high aspect ratio compared to other nanofillers [4].

Conductive ink is essential for flexible and stretchable printed circuits, ensuring defect-free and good adhesion between substrate and ink. This characteristic ensures circuitry can withstand bending, vibration, thermal, shock, and stretching loads [5]. Torsion is defined as the deformation of an object as a result of a torque or twisting force that has been applied. The process imposes the rotation of an object about its longitudinal axis, which induces shear tension on the object's cross-section [6]. Torsion testing consists of twisting a sample along an axis. It is an effective technique for determining properties such as the breaking angle, torsional shear stress, maximal torque, and shear modulus of a material or its interface for evaluating conductive inks' long-term durability and reliability. Cyclic bending and stretchability tests assess reliability performance, addressing issues like increased resistivity and lack of adhesion between ink and substrate [7]. Other than that, torsion cyclic tests are essential for evaluating conductive inks' long-term durability and reliability, especially those incorporating graphene nanoplatelets (GNPs). These experiments simulate real-world situations, allowing for the development of improved formulas that can withstand deformations without significant decreases in conductivity or mechanical strength [8]. The material is twisted repeatedly during this testing process. The studies replicate mechanical strains faced by conductive materials and inks, such as elongation and flexion. This form of testing enables understanding of the progressive degradation that occurs over time, which is not immediate but occurs gradually over usage [9].

Ageing is the gradual deterioration of materials, components, or systems caused by variables like mechanical strain, climatic conditions, and operational usage [10]. Ageing for data reliability refers to assessing how the performance of materials, components, or systems is affected by different stresses over time [11]. The information data on the ageing process is of utmost importance concerning the reliability of electronic materials, such as conductive inks, since it directly impacts their durability, lifespan, and safety over an extended period [12]. Accurate ageing data enables manufacturers and engineers to create durable goods and design appropriate maintenance schedules, assuring sustained performance and safety over time. Electronic materials deteriorating when ageing, including conductive inks utilized in diverse electronic devices, can significantly affect their dependability and efficiency [13]. Comprehending and examining failure and ageing mechanisms is essential and can enhance the design and maintenance of electronic storage systems, leading to improved dependability and safety [14].

This study aims to investigate the effects of ageing effects after enduring a cyclic torsional test on electrically high thermal GNP/Ag conductive ink. The outcome of this study provides insights into the electrical and mechanical properties of the ink over time, which is critical for developing more dependable and stable stretchable electronics.

2.0 Material and Methodology

2.1 Sample Formulation and Preparation

The preparation begins with a graphene nanoplates (GNP) hybrid formulation containing silver flake (Ag) for conductive ink manufacturing. The significant ingredients utilized in this experiment were 25μm GNP fillers, silver flake as an additional filler for the hybridization process, 1-butanol and terpineol as organic solvents used to produce a paste, and ethanol as a chemical solvent for the powder GNP hybrid production. Specific amounts of GNP were mixed with a solvent, such as ethanol, for plasma treatment or modified high-grade pure graphene so that GNP may be mixed with other materials, such as silver, in this formulation.

The GNP/Ag formulation began by dissolving 0.005g of 25 μm GNP powder in 5 mm of ethanol, followed by thorough mixing using an ultrasonic bath for 10 minutes. After sonicating for 10 minutes, 0.429g silver flakes were added to the GNP-ethanol solution and sonicated for another 60 minutes. Upon completion of the sonication process, the mixture of GNP and silver compound was subjected to stirring at a temperature of 70 °C and a rotational speed of 200 revolutions per minute (rpm) until the ethanol solvent undergoes evaporation. The dry solution was cured in an oven at 250 °C for one hour, then pounded into a fine powder to produce GNP hybrid paste. The hybrid GNP powder was weighed to determine the butanol-to-terpineol ratio. For each 0.52g of GNP powder, three (3) drops of butanol and three (3) droplets of terpineol were required. Weigh the compound to get the weight value of the estimator in the Thinky Mixer machine. Start the mixer for three minutes at 2,000 revolutions per minute (rpm). After the mixing procedure, store the GNP/Ag paste in a sealed container. Fig. 1 depicts the preparation process.

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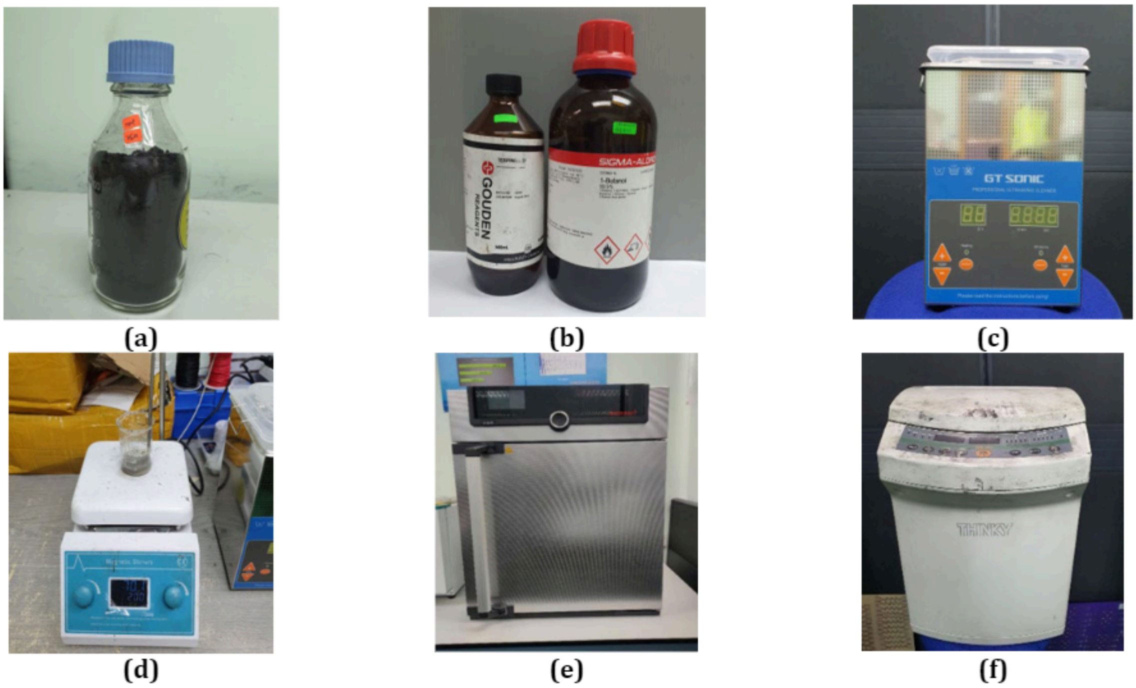


Fig. 1 *Preparation of GNP/Ag Conductive Ink paste. (a) GNP powder 25 μm size particle; (b) Ethanol and terpineol solvents; (c) GT Sonic machine for sonication process; (d) Magnetic stirrer: GNP/Ag heated at 70°C on a hot plate and stirred at 200 rpm; (e) GNP/Ag mixture dried in oven at 250°C for one-hour; (f) Thinky mixture machine to mix GNP-Butanol-terpineol for three minutes at 2,000 rpm*

2.2 Copper Substrate Preparation

The preparation of samples involves the application of copper-based materials in the form of strips, which serve as substrates made of copper. Copper was chosen as the substrate because of its high density, high thermal conductivity, and high specific heat capacity [15]. Substrate dimensions were 120 mm (length), 7 mm (width), and 0.1mm (thickness). While GNP paste dimensions were 3.5 mm (length), 3.5 mm (width), and 60 μm (thickness).

After sample preparation for the hybrid formulation, GNP/Ag paste was printed using a mesh stencil technique on copper substrates. The mesh stencil is 0.01mm thick. Each substrate was marked with a spacing of 20 mm from the center of each GNP conductive ink paste point as in Fig. 2. A small amount of the GNP/Ag ink was placed on a mesh stencil with a 3.5 mm x 3.5 mm grid and a squeegee was applied to transfer the ink to the substrate. This process is called the screen printing technique. The sample was cured in the oven for one hour at 250°C. Fig. 3 shows the processing steps for the sample preparation.

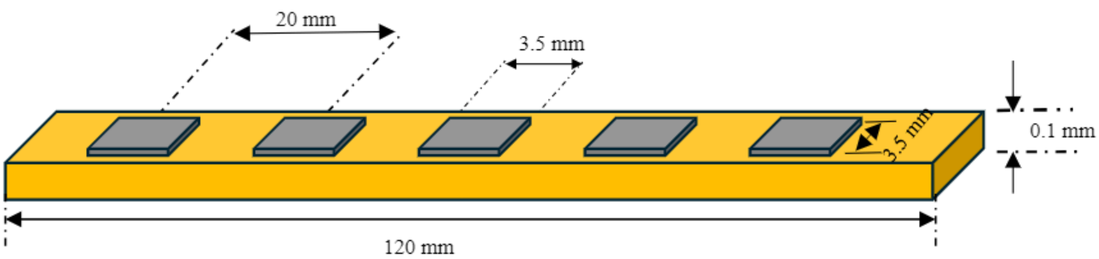


Fig. 2 *The GNP/Ag conductive ink on a copper substrate strip*

2.4 Resistivity Measurement Using Two-Point -Probe

Five designated monitoring spots (see Fig. 2) were identified as data collection points to measure the resistance. Resistance was measured at room temperature using a digital multimeter. Electrical resistivity is a characteristic that describes how effectively a substance resists the flow of electric current. Electric current flows efficiently through low-resistivity materials. Conductive ink electrical resistivity is defined as follows in Equation (1):

$$\rho = \frac{RA}{L} \tag{1}$$

where ρ is resistivity, R is the resistance recorded from the multimeter, A is the cross-sectional area, and L is the length. A two-point probe was used as the experimental apparatus for measuring the resistance value for every single substrate point. Zulfiqar et.al [16] used a two-point-probe multi-meter to measure the resistance of conductive ink. Data readings were taken by tapping at a specific position on each substrate point as in Fig. 4 and Fig. 5 shows the Two-Point probe apparatus being used.

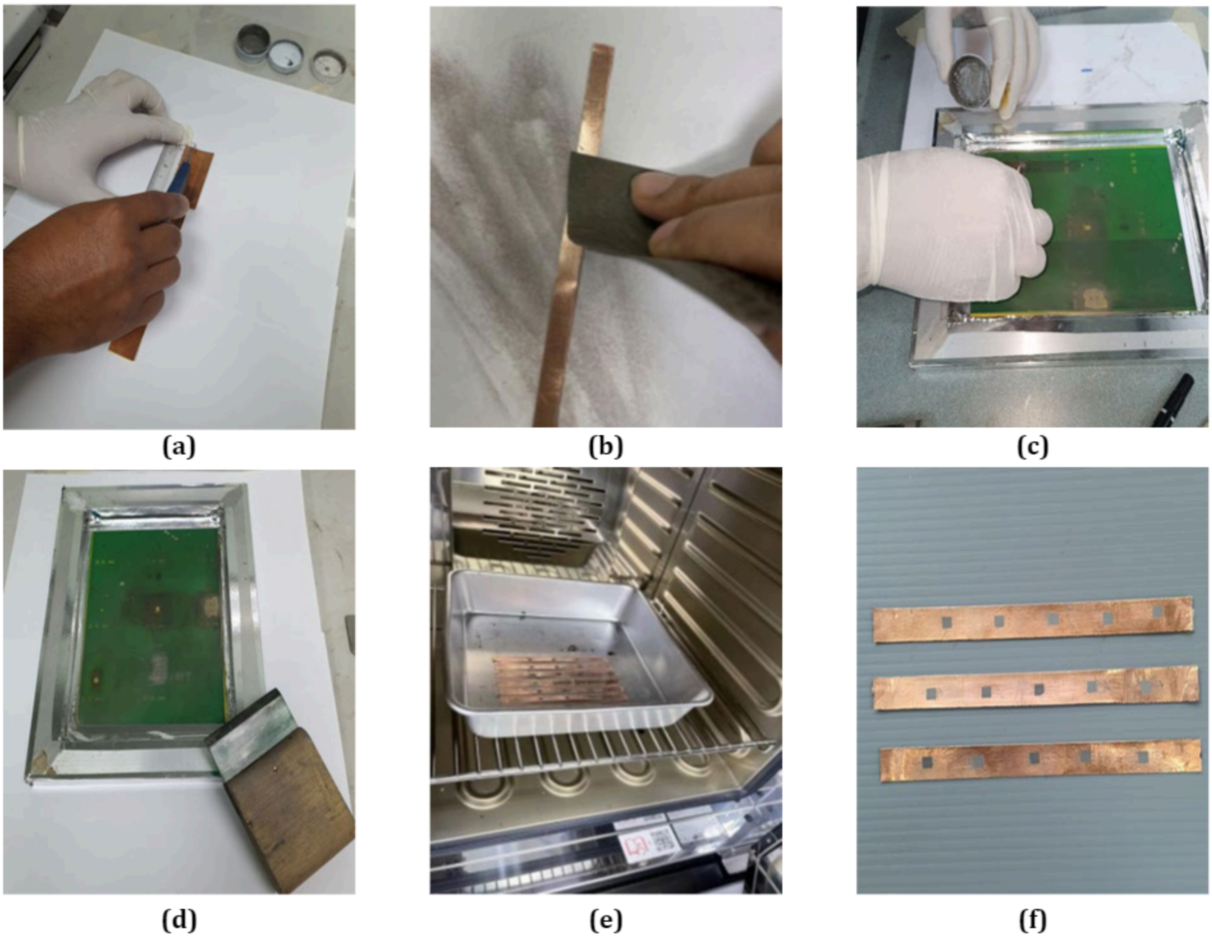


Fig. 3 *Preparation of Cooper substrate; (a) Cooper substrate was cut according to the size acquired; (b) Cooper was etched with sandpaper to remove carbon; (c) Printing process using mesh stencils and hybrid GNP/Ag paste was placed on the selected grid (3.5mm x 3.5mm); (d) GNP paste was applied to the strip by using scrapper; (e) GNP/Ag paste strip was put in the oven for curing process; (f) GNP/Ag paste strip ready for cyclic test*

Fig. 4 *Three points of printed GNP hybrid on copper substrates with observation positions for resistance measurement data collection using a two-point probe multimeter*

2.4 Torsion Cyclic Test

This test was conducted by using an in-house cyclic torsion test rig. The objective of this experiment is to determine the resistance and resistivity after several cycles of cyclic torsions. The GNP/Ag conductive ink substrates underwent cyclic rotation of 1000 cycles, 2000 cycles, and 4000 cycles. The strip was installed on the cyclic torsional test rig, in preparation for the experimental procedure. Two grippers created the samples' torsional motion; one remained stationary, whereas the other underwent iterative rotation. The twisting deformation was initially rotated clockwise and anticlockwise during the testing. The left strip was clipped on the rig's fixed end holder, while the strip's right end was hooked on the rig's twisting end gripper. A 12V DC motor powers the rack and pinion mechanism. Fig. 6 depicts the torsional test diagram.

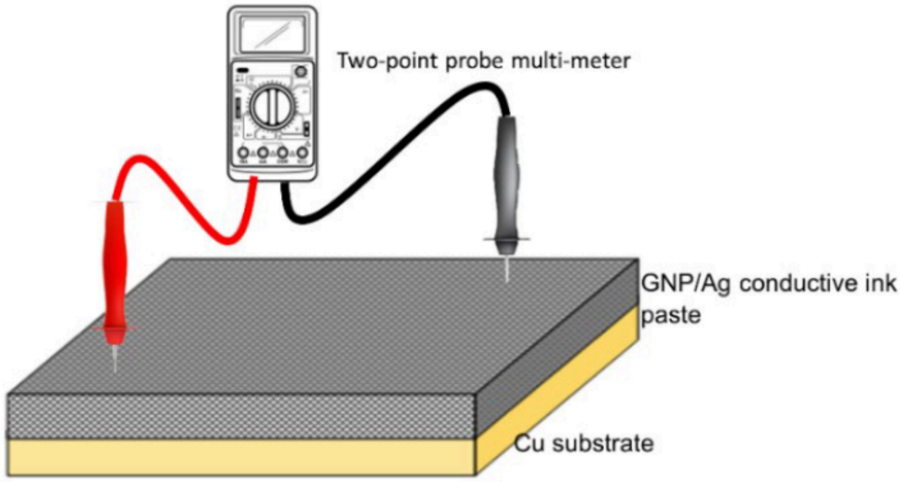


Fig. 5 *Resistance measured by a two-point probe*

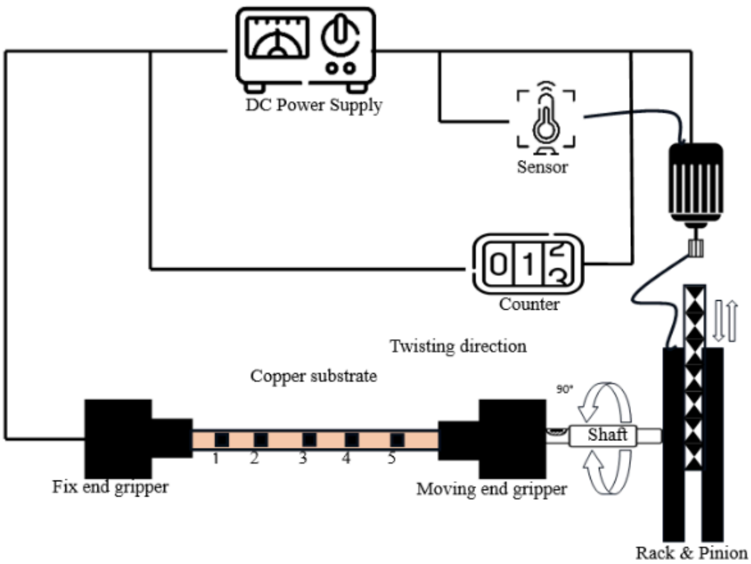


Fig. 6 *Cyclic torsional test diagram*

2.5 Ageing Observation Process and Data Measurement

A standard method for assessing the deterioration of sintered nanoparticles is by measuring their performance at elevated temperatures and then extrapolating these results to the temperatures of actual concern [17]. Gajadhur and Regulska, (2020) applied the Sun test + light chamber to accelerate the UV simulation and temperature elevation. Eventually, for this experiment, the strip was left exposed to the atmosphere for fifteen weeks at room temperature of 28°C (during office hours with air-conditioning) to 25°C (after office hours without air-conditioning). Measurements were collected once every week. Previous researchers, [10], [19], and [20] employed a four-point probe multimeter to measure electrical resistivity, while [7] applied Inductance, Capacitance, and Resistance (LCR) meter to measure inductance, capacitance, and resistance.

This experiment applied a two-point probe multimeter to measure the resistance at each of the five designated points; the outcome of the reading was subsequently documented. The resistivity value was calculated utilizing a formula implemented in the data record form using the Microsoft Excel software. The acquired data was subsequently visualized as a graph utilizing the Origin software. For this experiment, resistivity versus week graphs were constructed. The data was subsequently analyzed to determine the rate of resistivity reduction in conductive ink during the fifteen-week ageing process.

2.6 Microstructure Properties

Preparing a sample for SEM analysis using epoxy is imperative, especially for brittle or porous materials that require support to maintain their structure during inspection [21]. The technique started by embedding the sample in epoxy, which, after hardening, created a sturdy and stable matrix. At first, a small portion of the thin film of GNP/Ag conductive ink was cut out from the preferred spot. After that, the sample was cleaned to remove any contaminants. A rubber hose was cut in the middle to act as a holder, ensuring that the sample remained in the mold while epoxy was poured into it and allowed to solidify.

Then, the sample was grinded and polished to be as smooth as possible to avoid flaws that can impede SEM imaging [22]. The uncoated sample was later mounted to the SEM stub and placed inside the SEM chamber. The microstructure characteristics were studied to obtain particle distribution images of the GNP ink layer surface. The ink cross-section microstructure images were captured using JEOL JSM-5050PLUS/LV Emission Scanning Electron Microscopy (SEM) at an accelerating voltage of 5 kV.

3.0 Result and Discussion

Results and discussion were based on the data obtained from the carried-out experiment. Explanation and elaboration from these data focus on resistance and resistivity data gathered from multiple cyclic torsion tests (1000, 2000, and 4000 cycles) and followed by studies on the effect of ageing factors on 4000 cycles of torsion test strips. Both observations relate to each other as they reflect the outcomes of GNP/Ag conductive ink resistivity.

Table 1 *Resistance and resistivity data for 25μm GNP/Ag substrates for baseline and after 1000, 2000, and 4000 cycles of cyclic torsion test*

Cycles	GNP conductive ink paste	Average Resistance, (Ω)	Standard Deviation Resistance	Average Resistivity, (Ω.m)	Standard Deviation Resistivity
Baseline	1	0.53	0.06	1.867×10^{-03}	2.021×10^{-04}
	2	0.57	0.12	1.983×10^{-03}	4.041×10^{-04}
	3	0.63	0.06	2.217×10^{-03}	2.021×10^{-04}
	4	0.73	0.06	2.567×10^{-03}	2.021×10^{-04}
	5	0.53	0.06	1.867×10^{-03}	2.021×10^{-04}
1000 cycles	1	0.45	0.10	1.575×10^{-03}	3.500×10^{-04}
	2	0.48	0.15	1.663×10^{-03}	5.346×10^{-04}
	3	0.55	0.06	1.925×10^{-03}	2.021×10^{-04}
	4	0.80	0.00	2.100×10^{-03}	0.000×10^{-04}
	5	0.43	0.06	1.488×10^{-03}	2.021×10^{-04}
2000 cycles	1	0.70	0.06	2.450×10^{-03}	2.021×10^{-04}
	2	0.75	0.10	2.625×10^{-03}	3.500×10^{-04}
	3	0.78	0.06	2.713×10^{-03}	2.021×10^{-04}
	4	0.73	0.12	2.538×10^{-03}	4.041×10^{-04}
	5	0.78	0.06	2.713×10^{-03}	2.021×10^{-04}
4000 cycles	1	0.78	0.06	2.713×10^{-03}	2.021×10^{-04}
	2	0.80	0.06	2.800×10^{-03}	2.021×10^{-04}
	3	0.85	0.06	2.975×10^{-03}	2.021×10^{-04}
	4	0.80	0.12	2.800×10^{-03}	4.041×10^{-04}
	5	0.83	0.10	2.888×10^{-03}	3.500×10^{-04}

3.1 Results from Multiple Cyclic Torsion Test

Based on the experimental work conducted in this study, the values of average resistance and resistivity were observed and studied. The graph from the study consisted of the effect of cyclic torsion on GNP/Ag inductive ink substrate. GNP/Ag inks' electrical characteristics were assessed by measuring resistance and calculating resistivity on printed samples. Table 1 shows the average resistance and average resistivity values for GNP/Ag conductive ink strips after the cyclic torsional test.

Resistance and resistivity data obtained from the experiment were tabulated on resistance and resistivity versus cycles data as in Fig 2.8. The trend of resistivity as shown in Fig. 7 indicates that resistivity tends to increase with the number of cycles. The scatter of data points within each cycle category does not indicate a significant spread, suggesting a consistent response of the material's torsional property to resistivity changes within each cycle group. The average resistivity for baseline values from table, indicates consistencies at the initial value before the torsional cyclic test started. As for the 1000 cycles group, the resistivity values slowly show minimal changes in resistivity values. In contrast, the 2000 and 4000 cycles group exhibit incremental resistivity values which highlights the apparent impact of twisting cycle repetition on the conductive ink sample.

Twisting at high repetitive motion produces shear stress across the conductive ink strip, thus creating a deformation effect on each conductive ink point. Twisting produced shear, but the thin film's stress distribution was not uniform. Point three shows the highest resistivity among other ink points across all cycles. The phenomenon occurs at the most critical twist point, at the substrate's centre point. Point three withstands the most twisting effect from the torsion, followed by points four and five. Points two and one do not seem to be affected by the twisting motion. It could happen when the distribution from stress load decreases after point three, therefore minimizing the deformation effect on those two points.

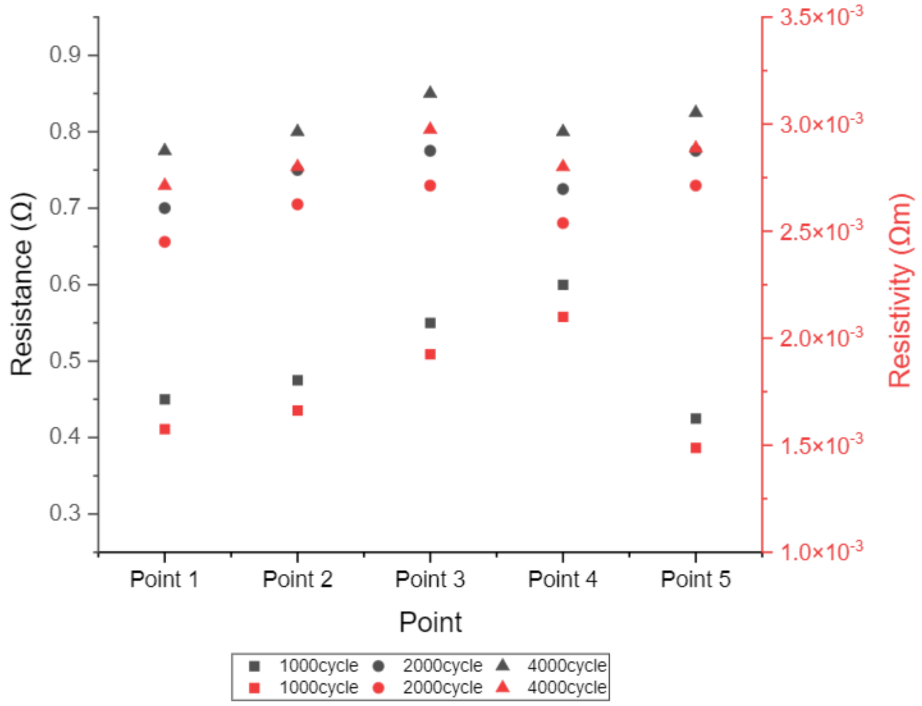


Fig. 7 *Resistance data from cyclic torsion test*

Thin films generally show instabilities and produce elaborate wrinkles when two clamped ends are twisted. Wrinkles appear at both ends of the film and move towards the centre, resulting in a localised area [23]. The data from the torsional cyclic test indicates that the resistivity increases as it endures more cycles, but the changes are not so significant, and it proves that the formulation of GNP/Ag conductive ink for this test able to withstand deformation and minimize the deformation impact due to fatigue from cyclic loading or the breakage of connections between the metallic grains. However further studies on ascending factors of resistivity values should be carried out to gain more insight regarding these issues. Several factors can influence the increasing value of resistivity in materials as they undergo repeated cycles or stress, including the sintering effect, [24] from the sample preparation phase, granular grains brittleness during stretching, bending, or cyclic repetition [25], crack formation [9] and breakage in conductive filler particles [26], which reduce carrier mobility. To identify the accurate factors contributing to the rise in resistivity, comprehensive investigations such as electron microscopy [27], X-ray diffraction, scanning electron microscopy [17], laser microscopy, or X-ray imaging [28] and other methods for characterising materials would typically be employed.

3.2 Resistance and Resistivity Data Measured from Ageing Effect from Torsion Test

In this second section, the results of the torsional cyclic test aimed at investigating the ageing effect on a GNP/Ag conductive ink were presented, together with photographic visuals of physical deformation of the GNP/Ag conductive ink condition during the 15-week observation period and resistivity measurement process. The torsional cycle test was designed to simulate the ageing process of the GNP/Ag conductive ink by submitting it to repeated torsional loads. The decrease in resistivity is a consequence of the ageing effect. Biermaier et al., (2021), categorized ageing into two distinct types: substrate ageing, which refers to changes in material performance, and functional ageing, which pertains to the deterioration of sensor behaviour, conductivity, and capacitance. The mechanical ageing test involves subjecting the material to repetitive treatments at a specific level of intensity. A visual evaluation of the physical deformation of the GNP/Ag conductive ink over the 15-week observation period revealed some significant changes. Initially, the GNP/Ag conductive ink appeared homogenous and densely packed, indicating a stable structure. However, as the test went on, the ink revealed indications of ageing, including apparent cracks and deformation. These changes suggest a deterioration in the ink's structural integrity, leading to a decline in conductivity and performance over time. To further understand the effect of ageing on the conductivity of the GNP/Ag conductive ink, resistivity measurements were taken weekly during the testing process.

The data acquired from the weekly measurements recorded with the universal two-point probe (ASTM F2750-16) are presented in Table 2. The unit for resistance is expressed in Ohms (Ω), while for resistivity is expressed in Ohmmeters (Ω.m). The precision of the decimal point has been adjusted to two decimal places to simplify the process of data recording. The baseline reading serves as a reference point for monitoring the changes in resistance and resistivity values due to the ageing process. The initial measurement indicates a progressive rise in the resistivity value from point one to point four. There is a decrease in resistivity value at point five. The highest value of baseline resistivity is noticed at point four, measuring 7.00×10^{-3} Ω.m. The trend exhibits a moderate inclination. From the first week to the fifteen weeks, it can be seen that the trend of resistance readings coincided with the trend of resistivity value readings.

Table 2 *Ageing resistance and ageing resistivity data for 25μm GNP/Ag substrates after torsion cyclic test*

		Average Resistance, (Ω)					Average Resistivity, (Ω.m)				
		Conductive Ink Point on Copper Substrate									
		1	2	3	4	5	1	2	3	4	5
Ageing Period	Baseline	0.53	0.57	0.63	0.73	0.53	1.87	1.98	2.22	2.57	1.87
	Week 1	0.78	0.80	0.85	0.80	0.83	2.71	2.80	2.97	2.80	2.88
	Week 2	0.83	0.48	0.63	0.45	0.60	2.89	1.67	2.19	1.58	2.10
	Week 3	0.83	0.45	0.50	0.45	0.50	2.89	1.58	1.75	1.58	1.75
	Week 4	1.10	0.73	0.87	0.83	0.90	3.85	2.57	3.03	2.92	3.15
	Week 5	1.20	0.97	0.87	1.03	1.13	4.20	3.38	3.03	3.62	3.97
	Week 6	1.27	1.10	1.07	1.07	1.17	4.43	3.85	3.73	3.73	4.08
	Week 7	2.00	2.50	2.43	2.43	1.50	7.00	8.75	8.51	8.51	5.25
	Week 8	0.93	1.20	1.83	0.93	1.33	3.27	4.20	6.42	3.27	4.67
	Week 9	0.93	1.53	1.47	2.00	1.40	3.27	5.37	5.13	7.00	4.90
	Week 10	0.93	0.70	0.57	1.57	1.43	3.27	2.45	1.98	5.48	5.02
	Week 11	1.17	1.23	0.97	1.67	1.63	4.08	4.32	3.38	5.83	5.72
	Week 12	2.00	1.27	2.77	4.10	1.30	7.00	4.43	9.68	1.03	4.55
	Week 13	0.47	0.63	0.93	1.13	1.33	1.63	2.22	3.27	3.96	4.67
	Week 14	0.33	0.23	0.83	1.27	1.40	1.17	8.17^{-4}	2.91	4.43	4.90
	Week 15	1.00	0.67	1.23	1.53	1.37	3.50	2.33	4.32	5.37	4.78

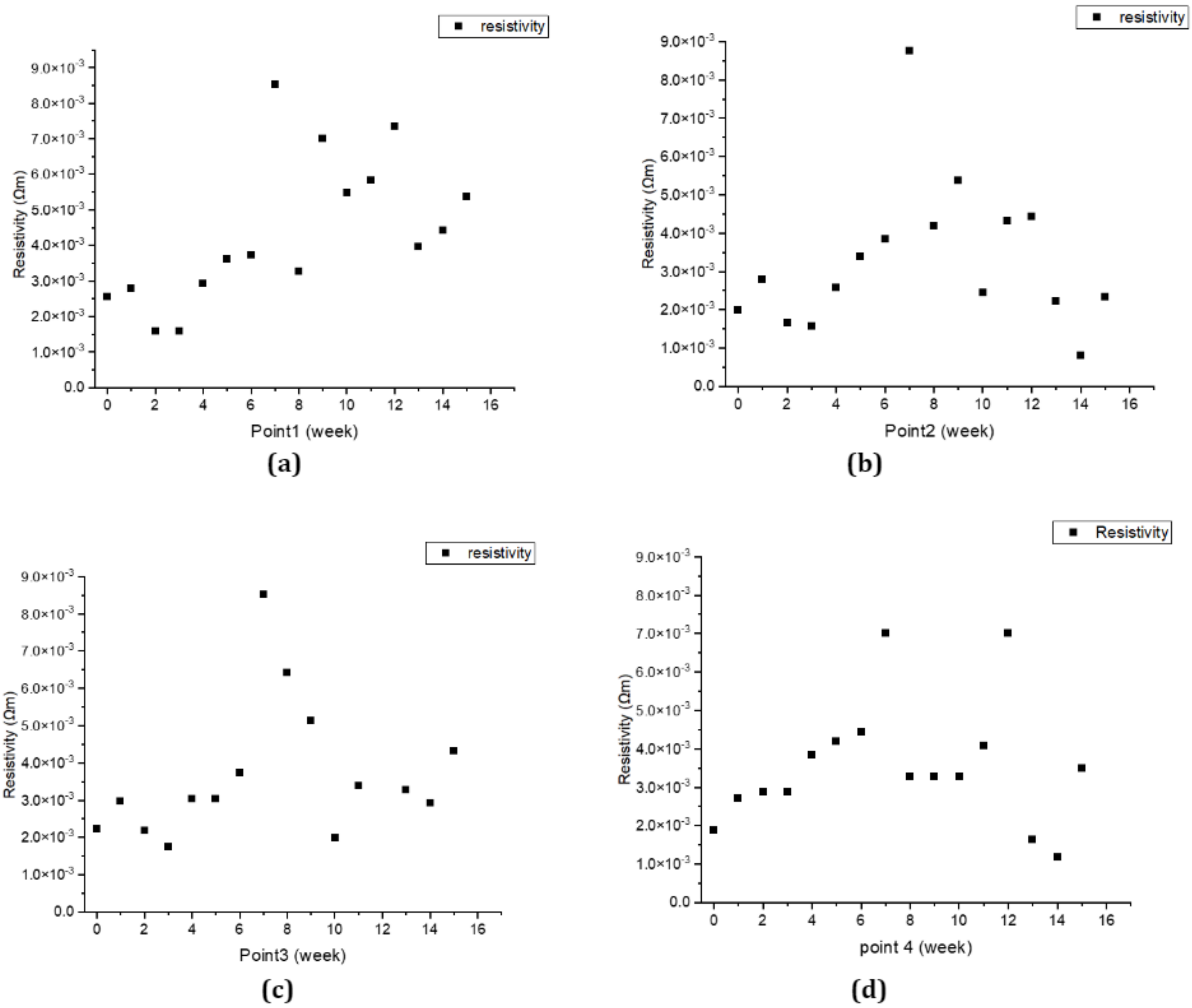
The initial hypothesis from this ageing experiment is the resistivity will show a gradual linear trend over time. Fig. 8 (exhibit the resistivity polarization for each observation point for an ageing period of 15 weeks. Generally, all points depict an incrementation pattern in resistivity. It indicates that ageing has taken place and resistivity has increased. During the first three weeks, the resistivity starts at a slow level and gradually increases. The damage does not begin instantaneously but rather accumulates over time. Raj et.al [17], acknowledged that the conductivity degradation in conductive ink related to ageing is a slow process. It may indicate the initial phases of the ageing process, during which the physical alterations may affect the material's conductivity. Fig. 9 shows the initial condition of the conductive ink substrate.

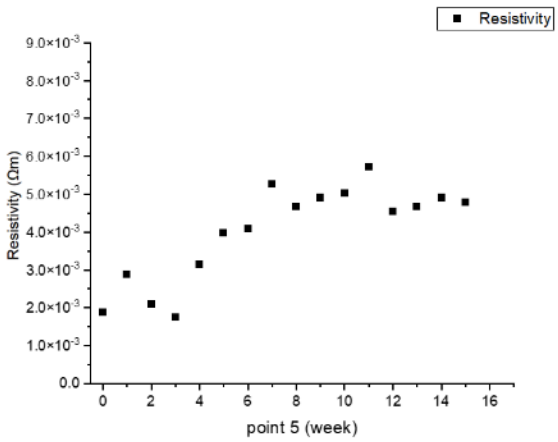
From week 3 to week 5, resistivity continues to rise but at a slower rate. As depicted in Fig. 10 (a), (b), and (c), each point of conductive ink has a minor, observable indentation. However, the ink remains utterly intact without any indications of fractures. A noticeable increase in resistivity from week 6 to week 9 suggests a faster ageing process. It indicates that the cyclic test performed on the inductive ink might start to produce cracks in the paste formation, resulting in conductivity path breakdown after 4000 cycles. Several possibilities can be counted for, including oxidation or scratching [17] on conductive ink that results in a breakage of conductive pathways [15]. As the ageing process continues, the ink shows clearer damage on the surface. Detrimentation on the conductive ink surface appears significantly in Fig. 10 (b) and Fig. 10 (c). Scratches are the main factor that leads to this outcome.

Scratches on the conductive paste surface are another possibility that contributes to the reduction in conductivity of this GNP/Ag paste. Scratching or damaging a conductive ink layer's surface can substantially impact its resistance and overall electrical performance. Overpressure from the tip of the Two-Point probe needle point created indentation which led to crack formation on the inks while obtaining the resistance value at each point. Fig. 11 displays the condition of indentation to the surface.

This action caused the ink to experience denting or crack and penetrate the Cu substrate and eventually disrupt the pathways by creating gaps, breaks, or discontinuities as in Fig 3.7. Gao et.al [24], proved that using a sharp indenter creates deeper scratches and causes more brittle failure. As a result, it also significantly affects the resistance of conductive ink. The scratching of the conductive ink layer may result in the creation of high-resistance points or contact discontinuities. These areas can impede the conductive flow or conductive pathway of electrons across the surface, leading to an increase in contact resistance. Elevated contact resistance contributes to higher overall resistivity. Scratching can initiate the formation of microcracks within the conductive ink layer, breaking of the 3D network formed by the conductive filler particles [29], loss of contact between different particles, an increase of the inter-filler distance, delamination of particles, reorientation of particles and decrease of the volume fraction of the filler material [26].

Scratched regions expose the underlying substrate and the conductive ink to environmental elements such as moisture, air, and pollutants. Exposing the ink to this can cause the metal particles within it to experience oxidation or corrosion, resulting in a deterioration of conductivity and an increase in resistivity as time passes. Influencing factors may also be due to the delamination of conductive layers, dissolution of conductive material, or transformation into a non-conductive material. Hence, determining the metal content is also an indicator of functional ageing.





(e)

Fig. 8 *Resistivity ageing data distribution separated by each GNP/Ag conductive ink point on Cu substrate strip for 15 weeks*

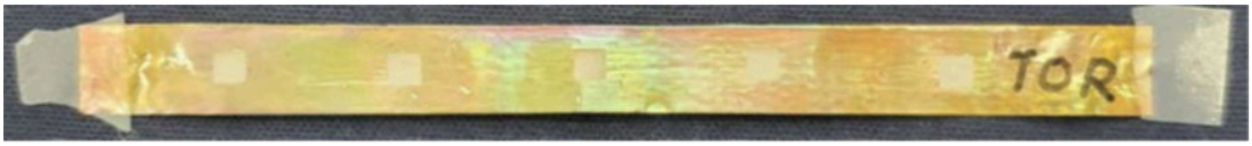
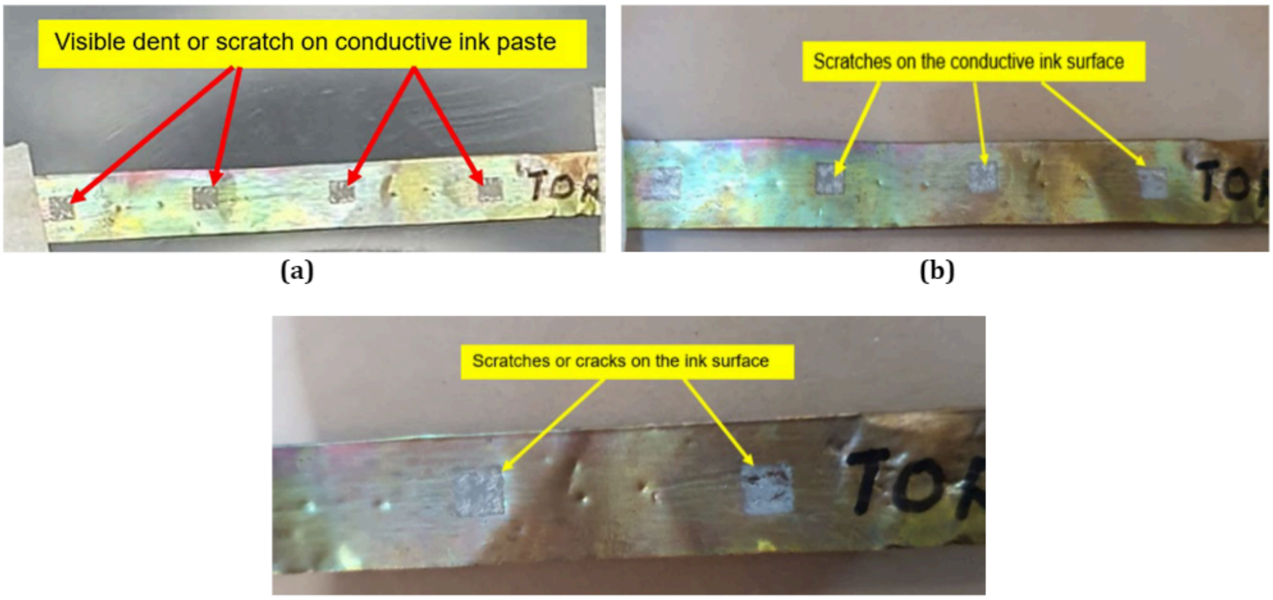


Fig. 9 *The GNP/Ag hybrid on copper substrates for cyclic torsion test*



(a) **(b)**

(c)

Fig. 10 *Dent on GNP/Ag paste on week three; Dent on GNP/Ag paste on week ten; Dent on GNP/Ag paste on week fifteen*

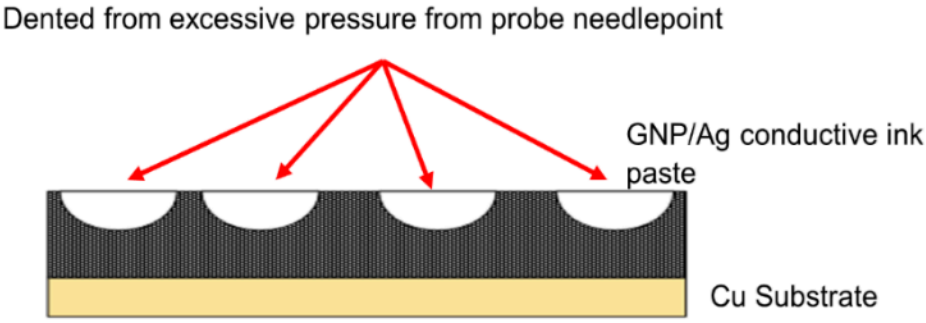


Fig. 11 *Dented effect on conductive ink paste*

3.3 Analysis of Morphology

The SEM image in Fig. 12 below displays a high-resolution depiction of the surface topography of the GNP/Ag conductive ink with 60 µm thickness. With the magnification of 2000x, an interconnected network of surface microcracks is clearly noticed, appearing as linear and branching patterns stretching across the ink layer's surface. The image ratifies the microcrack occurrence on a thin film surface. The microcracks exhibit various widths and depths, reflecting the distribution of different stresses and propagation along the strip. The microcracks are not only on the top layer but penetrate the copper substrate. Surface microcracks in GNP/Ag conductive ink are activated by the twisting effect of the cyclic process and the repetitive over-pressure indentation from the two-point probe tip. The appearance of microcracks indicates that although the GNPs enhanced flexibility and strength, their agglomerations can create regions of high stress, especially in a matrix containing a different conductor, such as silver [30]. The imperfections lead to an enlarged particle gap that facilitates electron flow to the crystallographic misalignment in the graphene lattice with various grains [31]. The previous study from [32] also supports these similar findings. Therefore, it decreases the electrical conductivity of the ink. Hence it increases the resistivity value. However, the ink is still intact and conducts electricity. This indicates that the lower layers may maintain their structural integrity. Additionally, the cracks are mainly found at the edges of the GNP/Ag conductive ink point, indicating cracks occurred in the defect area.

This experiment outcome showed a significant probability that ageing occurred during the whole process due to data obtained from the experiment and the loss of resistivity in the GNP/Ag conductive ink. However, ageing was not the main factor related to this decreasing value. Other factors usually linked to ageing such as exposure to high temperature environments [17]; [33]; elevated temperature exposure [34], and exposure to direct sunlight [18], which were not affected to this condition.

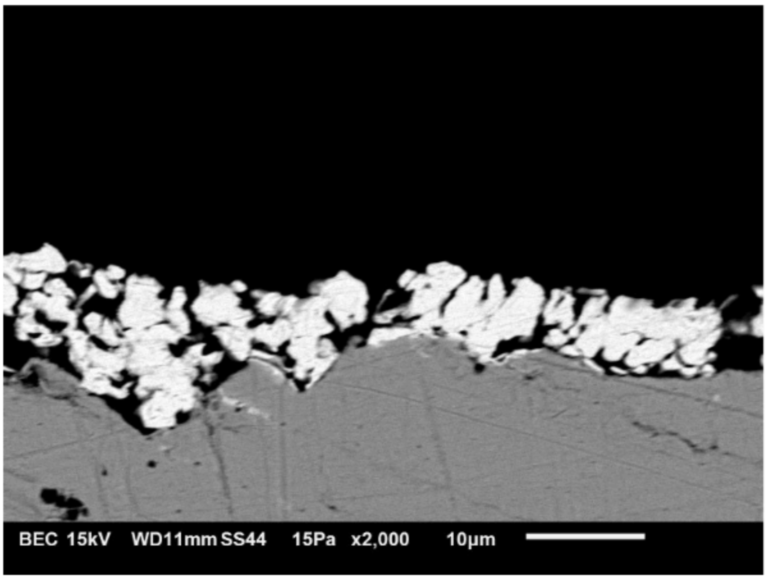


Fig. 12 *Surface microstructure image of GNP/Ag conductive ink with 60µm thickness*

4. Conclusion

The experiment successfully assessed the effects of ageing on a conductive ink based on a hybridization epoxy with high thermal conductivity. After 4000 cycles of torsional testing, the conductive ink demonstrated minimal resistance and a gradual increase in resistivity. Repeated twisting on a thin film, particularly in the study of conductive materials such as GNP/Ag conductive ink, may result in different mechanical and material alterations, impacting its electrical characteristics. The research findings indicated that the electrical conductivity of the conductive ink was not hugely affected by torsional cyclic twisting, thus proving that the ink was able to be electrically conducted even though deformation occurred and continues to operate at its best level. Throughout fifteen-week ageing test conducted on the thin film unveiled a significant increase in resistance. Over time, the samples underwent an ongoing ageing process. An investigation was being conducted into the sudden rise in resistance value, considering the sample had been preserved in a controlled environment devoid of excessive temperature exposure or accelerated ageing. The primary factor of this specimen's observed conductivity decline can be attributed to significant surface abrasion and indentation or cracking on the inductive ink. SEM morphological image ratified the conditions above. The electric current was disrupted by scratches, resulting in a progressive rise in resistance. To obtain accurate resistivity data, the researcher must replicate the experiment multiple times and exercise caution when using the two-point probe to record readings on the conductive ink points. Researchers should implement preventive measures during resistivity collection to prevent the inductive ink from scratching or breaking.

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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 contribution to the paper as follows: ***study conception and design****: Muhammad Arieef Hussain, Mohd Azli Salim;* ***data collection****: Nor Azmmi Masripan;* ***analysis and interpretation of results****: Muhammad Arieef Hussain, Adzni Md. Saad;* ***draft manuscript preparation****: Muhammad Arieef Hussain, 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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