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Influence of terpinol-butanol composition on electrical properties and ageing factor behaviour of hybrid conductive inks

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ABSTRACT

Hybrid conductive inks based on graphene nanoplatelets (GNP) and silver (Ag) are promising for printed electronics where electrical reliability under mechanical loading is required. This study evaluates the influence of the terpinol (T) to butanol (B) solvent ratio on the electrical behaviour and ageing response of hybrid GNP/Ag conductive ink. Three formulations, 5T:10B, 10T:10B and 15T:10B, were screen printed on copper (Cu) substrates and cured at 250 °C for one hour. Cyclic torsional loading at ±90° was applied from 1000 up to 4000 cycles, followed by room temperature ageing for five weeks. Resistance and resistivity increased with torsional cycling for all formulations. The most pronounced change occurred beyond 2000 cycles. The 10T:10B formulation exhibited the lowest resistivity after 4000 cycles, while the 5T:10B and 15T:10B compositions showed higher degradation associated with solvent ratio differences in filler distribution. During ageing, only minor resistivity changes were observed. No cumulative degradation was detected at room temperature. These results indicate that a balanced terpinol to butanol ratio supports a conductive network that is resilient to cyclic torsion and short-term environmental exposure. The 10T:10B solvent ratios provide the most favourable electrical stability.

Keywords: *Graphene nanoplatelets; Silver; Hybrid conductive ink; Terpinol–butanol ratio; Electrical conductivity; Ageing behaviour; Flexible electronics.*

1. INTRODUCTION

Solvents play an important role in keeping the appropriate viscosity in the formation of graphene nanoplatelet (GNP) and silver (Ag) conductive ink formulations. Each solvent is determined by the device application and the printing method. Solvents used include N-dimethylformamide (DMF), N-methyl-2-pyrrolidone (NMP), dimethyl sulfoxide (DMSO), terpinol, ethanol, isopropyl alcohol, ethylene glycol, and glycerol [1]. Most solvents pose a risk to human health and the environment. High annealing temperatures are typically required for conventional conductive ink to achieve sufficient electrical conductivity, which limits its applicability on flexible substrates. In contrast, alternative solvent systems with lower toxicity profiles have been explored to reduce processing risks and to avoid reliance on excessively high annealing temperatures. In this work, the term "flexible electronics" refers to the mechanical deformation mode imposed on the printed conductive ink rather than implying compatibility with low-temperature polymer substrates. Research has been conducted to create low-toxicity conductive inks that can operate effectively at lower annealing temperatures for flexible electronics applications [2]. Solvent is one of the main components for conductive inks [3]. Butanol is effective in enhancing the dispersion of nanoparticles and controlling the drying process. Terpinol provides a low toxicity characteristic. A good solubility combination with Ag compounds and improved stability [4]. Terpinol is also effective in dispersing and stabilising GNP [5-6]. Using these solvents will help improve the environmental safety of the manufacturing process and support sustainable manufacturing in electronics [7]. Viscosity is a key parameter in printed conductive ink because it governs ink transfer during screen printing and the continuity of the deposited ink. Ink viscosity influences stencil filling behaviour [8], edge definition and the ability of the ink to form a continuous conductive layer without excessive spreading or break-up [9]. Variations in viscosity are also reflected in particle mobility during drying, which affects the filler's packing density and junction formation within the printed ink [10]. As a result, viscosity changes associated with solvent composition are reflected in print quality as well as in the electrical resistance measured after curing and cyclic torsional loading.

Cyclic torsion testing determines the fatigue and service life of a material subject to repeated twisting loads. The test is widely applied in the automotive and aerospace industries [11]. Resistance and resistivity of conductive materials, such as hybrid GNP/Ag conductive ink, indicate stability under mechanical stress [12]. When assessing the performance of conductive inks under torsional loading, resistance and resistivity values reflected the effect of deformation and fatigue on electrical properties. For this test, one end of the sample is secured during the test, while the other end is rotated to produce shear stress. The test generates data concerning the fatigue life and shear strength of the sample [13-14]. Material ageing occurs when a material's performance changes over time as a result of physical, chemical or mechanical influences that may affect its performance and lifespan [15]. Material ageing can have a significant impact on the long-term stability of conductive inks, particularly under temperature, humidity and strain conditions [16]. In this research, the effects of terpinol-butanol ratios (5T:10B, 10T:10B and 15T:10B) on the resistance and resistivity of hybrid GNP/Ag conductive inks subjected to cyclic torsion were examined. The ageing behaviour of the inks was also evaluated in terms of electrical resistance at room temperature as a function of the solvent composition. An understanding of these relationships is necessary to ensure the reliability and durability of the conductive inks employed in flexible electronic devices [17].

2. MATERIALS AND METHOD

2.1. Materials

In this research, GNP (particle size of 25 µm and a surface area of 150 m^2/g) and Ag flakes (10 µm, ≥99.9% trace metals basis) and Ag acetate (SA) (99.99% trace metals basis) are used. Ethanol (denatured ethanol 99%), terpinol (~65% α, ~20% γ, ~10% β) and butanol (99.9%) were acquired from Merck KGaA, Darmstadt, Germany.

2.2. Sample Formulation and Preparation for Different Ratios of Terpinol

The preparation of the hybrid GNP/Ag conductive ink began with the preparation of ink containing GNP, Ag, SA and solvents (ethanol, butanol, and terpinol). Ethanol acts as a dispersion medium. It increases the viscosity and surface tension of the printing ink. The formulation procedures began with dissolving 0.5g of GNP powder was dissolved in 50ml of ethanol and ultrasonically mixing for 10 minutes. Then 0.429g of Ag flakes and 0.042g of SA were added to the GNP-ethanol solution and sonicated for 60 minutes to achieve a uniform dispersion. Complete removal of ethanol was ensured through controlled thermal processing rather than direct chemical analysis. The solution was stirred at 70°C at 200rpm, until a dry solid state was obtained, indicating solvent evaporation. The dried solution was then cured at 250°C for an additional hour to eliminate any residual solvent from the ethanol further. The cured powder was then ground into a fine powder to obtain a GNP/Ag hybrid powder. This curing step was applied during the powder preparation stage to remove residual solvent and does not represent a post-print curing process on a flexible substrate. This hybrid powder was then combined with terpinol and butanol to produce a conductive paste. For each 0.52g GNP powder, 10 drops of terpinol and butanol are used as the baseline ratio. Additional pastes were prepared with different terpinol-to-butanol (T:B) ratios of 5T:10B, 10T:10B and 15T:10B to test how the solvent's composition affects the properties of the conductive ink. The detailed formulations of the hybrid GNP/Ag conductive ink are presented in Table 1. The compounds were then mixed using a Thinky mixer at 2000 rpm for 3 minutes. The compound was placed into sealed containers to preserve the homogeneity of the mixtures for this testing.

Table 1. T:B mixing ratio

Sample	Powder				Organic Solvents	
	GNP (g)	Ag (g)	SA (g)	Ethanol (ml)	T (drop)	B (drop)
5T-10B	0.5	4.292	0.42	50	5	10
10T -10B	0.5	4.292	0.42	50	10	10
15T-10B	0.5	4.292	0.42	50	15	10

2.3. Preparation of Copper Substrate

The preparation of samples involves applying copper (Cu)-based materials in the form of strips, which serve as Cu substrates. Cu was selected as it was acknowledged for its high density, thermal conductivity and specific heat capacity [18]. The dimensions of the Cu substrate were 120mm (length), 7mm (width) and 0.1mm (thickness), and the GNP/Ag paste was 3.0mm (length), 3.0mm (width) and 60µm (thickness). The substrates were lightly abraded using sandpaper to remove surface oxide and contaminants, followed by cleaning with acetone, drying, and storage in a glass desiccator to minimise moisture uptake and re-oxidation before printing. After the hybrid formulation samples were prepared, GNP/Ag paste was printed onto the Cu substrates using a mesh stencil printing technique. The thickness of the mesh stencil measures 0.01mm. As illustrated in Figure 1, each Cu substrate was designated with a spacing of 20 mm from the centre of each GNP conductive ink paste point.

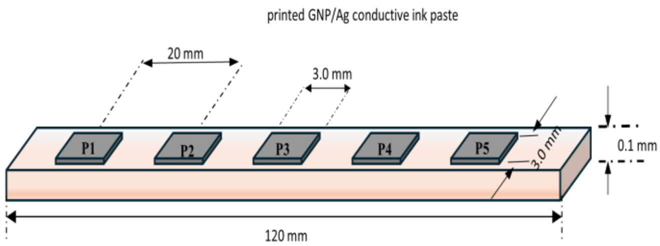

Figure 1. Printed GNP/Ag conductive ink paste on a Cu substrate

2.4. Resistivity Measurement using Two-Point Probe

Five designated monitoring spots (Figure 1) were identified as data collection points for measuring resistance. Resistance was measured at room temperature using a two-point probe 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. The resistance measurement was performed according to IEC 62899-202 guidelines. Conductive ink electrical resistivity is defined as follows in Equation 1:

$$\boldsymbol{Resistivity, \rho} = \frac{\boldsymbol{R}}{\boldsymbol{L}} \times \boldsymbol{A} \tag{1}$$

where R is the resistance, L is the length of the current path, A is the area of the conductive path and P1-P5 represents the test sample, a printed hybrid GNP/Ag conductive ink on the Cu substrate. The given formula to calculate the area of the test cross-section was as Equation 2:

$$\boldsymbol{Cross\ Section\ Area, A} = \omega \times \mathrm{L} \tag{2}$$

where multiply the length (L) of the conductive strip or patch by the applied ink's uniform thickness (ω). For these experiments, the thickness of the ink utilised was 60µm. Figure 2 illustrates the dimensions of the calculated GNP/Ag conductive ink paste cross-section area.

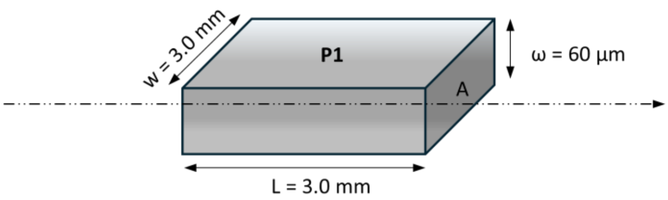

Figure 2. Illustration of the cross-sectional area of a hybrid GNP/Ag conductive ink paste

2.5. Cyclic Torsional Test

The cyclic torsion test utilised an in-house test rig to evaluate the resistance and resistivity of hybrid GNP/Ag conductive ink substrates subjected to repeated torsional cycles. The aim was to assess how resistance and resistivity changed after 1000, 2000, and 4000 torsion cycles. The hybrid GNP/Ag conductive ink substrate strip was securely clamped to the test rig using two grips that controlled torsional deformation. One gripper remained static, and the second gripper repeatedly rotated. It caused the strip to twist. The rotation alternates between clockwise and counterclockwise through each cycle of the test. The left end of the strip was tied to the static holder on the test rig, and the right end was tied to the rotating gripper. The torsional actuator consisted of a 12V DC motor that powered a rack-and-pinion system to provide consistent rotation to the torsional mechanism. The sample was subjected to a torsion range of ±0° to 90° at 100 rpm, as per ASTM A938-18 and ASTM E2207-15.

2.6. Ageing Analysis

This ageing test analyses the ageing behaviour of hybrid GNP/Ag conductive inks with different terpinol-to-butanol solvent ratios: 5T:10B, 10T:10B, and 15T:10B. The analysis was carried out after 4000 torsional cycles. Measurements were conducted over five weeks under room conditions at 25°C and 68% relative humidity. The Two-Point probe method, a standard technique for measuring electrical properties, was adopted in accordance with IEC 60068-2 guidelines. The purpose was to assess the stability of resistance and resistivity over time and to identify the solvent ratio that provides the most stable long-term performance.

3. RESULTS AND DISCUSSIONS

3.1. Electrical Properties under Cyclic Torsion

The results obtained in Figure 3 explain how the electrical properties of the hybrid GNP/Ag conductive ink vary based on different proportions of terpinol to butanol (5T:10B, 10T:10B, 15T:10B). All the samples were cured before being subjected to cyclic torsion testing until 4000 cycles. Resistance and resistivity both increased with the number of cycles, indicating that the structure of the conductive network is changing as it is subjected to these cycles. The rate at which these values changed depended upon the composition of the solvents. Sample 5T:10B followed with $2.07\text{x}10^{-5}$ Ω.m. When the number of cycles reached 4000, sample 10T:10B exhibited the lowest resistivity at $2.20\text{x}10^{-5}$ Ω.m., while the 15T:10B sample exhibited the greatest amount of resistivity at $2.73\text{x}10^{-5}$ Ω.m. These findings demonstrate that the mechanical strength of hybrid ink depends upon the ratio of the solvents present in it. The solvent composition plays a critical role in controlling the rheological behaviour, dispersion stability of conductive fillers, and evaporation dynamics during the curing process. A well-balanced solvent ratio promotes uniform dispersion of conductive particles such as graphene nanoplatelets and metallic fillers, which facilitates the formation of a continuous conductive network within the cured film. In addition, the solvent system affects the wetting behaviour of the ink on the substrate, influencing interfacial adhesion and film uniformity. If the solvent ratio is not optimised, rapid or uneven evaporation may occur, leading to defects such as microvoids, weak particle–particle contacts, and poor film densification. These microstructural imperfections can significantly reduce the mechanical integrity and durability of the printed conductive layer. Therefore, careful control of the solvent composition is essential to ensure proper film formation, enhance interparticle bonding, and ultimately improve the mechanical robustness and reliability of the hybrid conductive ink under mechanical and environmental loading conditions.

3.1.1 5T:10B Ratio Composition

The resistance and the resistivity graph of 5T:10B in Figure 3 shows an upward trend under cyclic torsional loading. The increase in resistance and resistivity develops gradually with increasing torsional cycles. Changes remain limited at lower cycle counts to 1000 cycles, followed by a steady rise between 1000 and 2000 cycles. A more pronounced increase is observed beyond 2000 cycles, indicating cumulative degradation of conductive junctions as mechanical loading continues. At 4000 torsional cycles, the total average resistivity of the 5T:10B formulation reached $27.3\text{x}10^{-6}$ Ω.m, corresponding to a total average resistance of 0.46 Ω. Figure 3 presents the data as a plotted graph, where total average values are shown with error bars representing the corresponding standard deviation.

Repeated mechanical cycling affects the hybrid conductive ink when partial debonding or slippage occurs between individual conductive particles. Such behaviour is commonly observed when hybrid inks are subjected to repeated bending or torsional deformation [19]. Progressive separation of neighbouring Ag particles weakens electrical connectivity within the conductive network and increases the resistance associated with the Ag to Ag junctions. As junction resistance increases, the overall electrical conductivity of the hybrid ink decreases cumulatively without experiencing sudden failure.

The response of the 5T:10B formulation under cyclic torsion is closely linked to the solvent composition used during ink preparation. Butanol ($C_4H_{10}O$), with a boiling point of 117.7°C and a dipole moment of 1.66 D, evaporates relatively rapidly during curing. This rapid evaporation

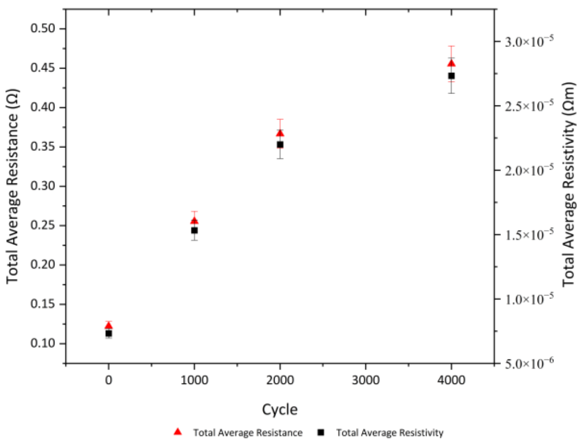

Figure 3. Terpinol to butanol ratio 5T:10B

promotes faster ink drying but can limit extended particle rearrangement, increasing sensitivity to mechanical deformation during subsequent cyclic loading. In contrast, terpinol ($C_{10}H_{18}O$) has a higher boiling point of approximately 219°C, resulting in a slower evaporation rate that supports improved dispersion of conductive fillers. Differences in solvent structure and polarity influence their interactions with the Ag nanoparticles and govern the quality of filler dispersion within the hybrid ink. Solvents with higher polarity promote nanoparticle dispersion by reducing surface charge repulsion and aggregation tendencies [20- 21].

Within the 5T:10B formulation, the good chemistry of the combined use of terpinol and butanol produces a miscible solvent system that supports an initially uniform ink state during drying. However, the relatively higher butanol content favours faster solvent removal. This accelerates the drying process and increases the likelihood of local structural adjustment under cyclic torsion. Together, these effects explain the gradual yet increasingly pronounced rise in resistance and resistivity observed at higher cycle counts. Here, the term "good chemistry" refers to their complementary and miscible behaviour. This compatibility supports uniform filler dispersion without implying any chemical reaction between the solvents.

3.1.2 10T:10B Ratio Composition

Compared with the 5T:10B formulation, the 10T:10B composition exhibits a more reliable electrical response during the early stages of cyclic torsional testing, as shown in Figure 4. Resistance remains relatively stable during the initial loading stage, increasing only slightly from 0.12 Ω to 0.14 Ω as cycling progresses to 2000 cycles. Beyond this point, a more pronounced increase in resistance is observed, reaching 0.37 Ω at 4000 cycles. A similar trend is recorded for resistivity. Initial resistivity values remain close to 7.33×10^{-6} Ω.m up to 2000 cycles, followed by a sharper rise to 2.20×10^{-5} Ω.m at 4000 cycles. This behaviour indicates that electrical degradation in the 10T:10B formulation develops gradually and becomes more apparent only after extended mechanical loading.

The comparatively stable response observed during the early cycle stages reflects the influence of solvent balance on filler distribution and junction integrity. In the 10T:10B formulation, the combined effects of terpinol and butanol promote a uniform dispersion of conductive fillers during curing. Terpinol provides a slower evaporation rate, allowing sufficient time for particle rearrangement, while butanol contributes to consistent ink formation through controlled volatility [22]. This balance supports the formation of a compact conductive layer with limited agglomeration and reduced void formation, which helps maintain electrical continuity during the initial phases of torsional deformation. Previous studies have shown that inks with appropriate viscosity and solvent balance tend to exhibit improved filler distribution and fewer micro voids, which contribute to enhanced electrical stability under mechanical stress [23]. The performance of conductive ink, particularly that incorporating the GNP, is highly dependent on the solvents used in the formulations [24].

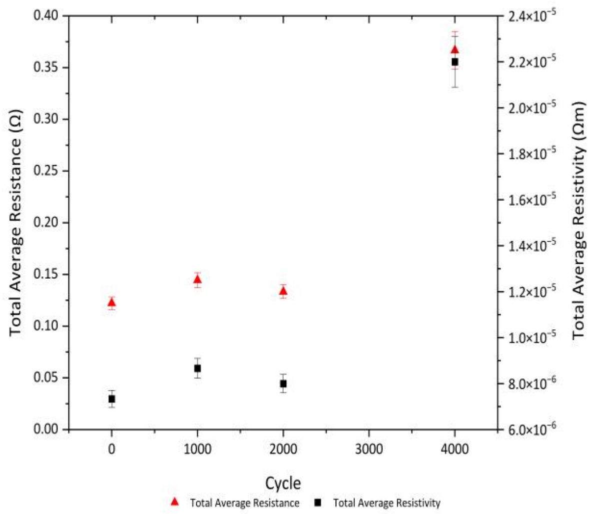

Figure 4. Terpinol to butanol ratio 10T:10B

As torsional cycling continues, the accumulation of mechanical strain begins to affect the conductive network. After prolonged loading beyond 2000 cycles, repeated deformation promotes local disruption of conductive contacts, including partial separation of the Ag particles and the gradual loss of effective percolation pathways. These changes manifest as a more pronounced increase in resistance and resistivity at higher cycle counts, rather than as an abrupt failure. The delayed onset of this behaviour distinguishes the 10T:10B formulation from the 5T:10B composition, where degradation becomes evident at earlier stages.

Overall, the electrical response of the 10T:10B formulation reflects a balance between dispersion stability and mechanical durability. The solvent composition supports a well-distributed conductive network that resists degradation during the early stages of cyclic loading. Extended torsional loading eventually leads to progressive fatigue within the conductive layer. This behaviour positions the 10T:10B formulation as a reference composition. It combines stable early cycle performance with predictable and gradual degradation under prolonged mechanical strain. This relationship provides a clear link to the ageing behaviour discussed in the subsequent section.

3.1.3 15T:10B Ratio Composition

The 15T:10B formulation exhibits a higher initial electrical resistance and resistivity compared with the other solvent ratios, as shown in Figure 5. At zero cycles, the resistance is measured at 0.24 Ω with a corresponding resistivity of 1.47×10^{-5} Ω.m. As torsional cycling proceeds, both parameters increase, reaching a resistance of 0.46 Ω and a resistivity of 2.73×10^{-5} Ω.m at 4000 cycles. This response reflects a comparatively less uniform conductive network established during curing, which is more sensitive to mechanical deformation.

During the early stage of cyclic loading, changes in resistance and resistivity are moderate. From zero to 1000 cycles, resistance and resistivity increase by 8.33% and 4.08%, respectively. As cycling progresses beyond 1000 cycles, a more pronounced rise is observed, with resistance and resistivity increasing by 53.85% and 56.86% at 2000 cycles. Beyond this point, the rate of increase slows as the cycle count approaches 4000 cycles. This trend indicates that electrical degradation in the 15T:10B formulation develops primarily during the intermediate cycle range, followed by a gradual transition toward a more stable response at higher cycle counts.

The electrical behaviour of the 15T:10B formulation is closely linked to the high terpinol content in the solvent system. Terpinol is a viscous and slow-evaporating solvent that promotes the formation of thicker ink layers during curing. This condition restricts particle mobility. It also encourages local clustering of conductive fillers, leading to spatial variations in filler density [25].

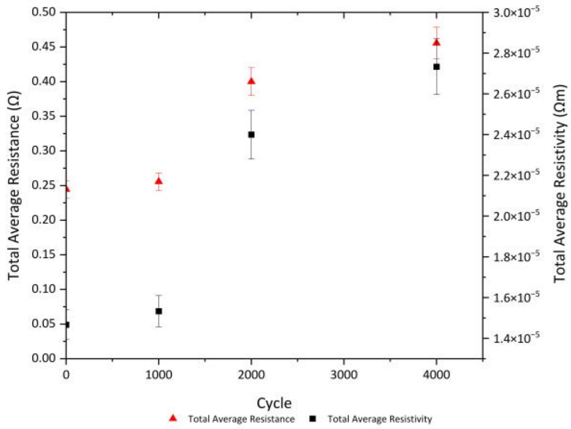

Figure 5. Terpinol to butanol ratio 15T:10B

Such heterogeneity weakens the continuity of the percolation network and increases the sensitivity of conductive junctions to mechanical strain [26]. During curing, the extended drying time further limits article rearrangement, which contributes to the formation of locally stressed regions within the conductive layer.

As torsional cycling continues beyond 2000 cycles, the rate of resistance increases. This behaviour suggests that repeated mechanical deformation allows partial rearrangement of conductive contacts. This rearrangement reduces the rate of further junction degradation. The presence of the GNP supports electrical continuity by providing additional conductive pathways that help maintain charge transport even when the Ag particle contacts are locally disrupted [27]. This redistribution of current reduces the likelihood of abrupt electrical failure, although it does not fully compensate for the structural heterogeneity introduced by the high terpinol content.

Overall, the 15T:10B formulation demonstrates a distinct electrical response characterised by higher initial resistivity. It also exhibits a pronounced increase in resistivity during the intermediate stages of cyclic loading. A gradual stabilisation occurs at higher cycle counts. The excessive terpinol content limits dispersion uniformity and promotes clustering, which results in a conductive network that is less homogeneous than that of the 10T:10B formulation. This behaviour explains the increased sensitivity to torsional loading while remaining consistent with the controlled ageing response observed in the subsequent section.

3.1.4 Comparative Interpretation

Comparing the formulations highlighted the dominant effect of the solvent balance on the resilience of the hybrid GNP/Ag conductive ink. The 5T:10B formulation exhibits a gradual and distributed increase in resistance, which is associated with faster solvent evaporation and moderate dispersion uniformity. In contrast, the 10T:10B formulation shows the most stable electrical response during early and intermediate torsional cycles, reflecting a more uniform conductive network established during curing. These findings align with previous reports that both the polarity and evaporation rate of solvents are key factors influencing the distribution of fillers and the formation of conductive pathways in hybrid inks [28]. The 15T:10B formulation presents higher initial resistivity and a more pronounced increase beyond 1000 cycles, which is consistent with restricted particle mobility and localised clustering linked to higher terpinol content.

Across all compositions, the evolution of resistance with increasing cycle number reveals a consistent trend. The pattern shows that the way the solvents regulate the distribution of fillers plays a crucial role in determining the electrical behaviour. This regulation of filler distribution influences when degradation begins during mechanical loading. It also governs how this degradation develops as the material is subjected to torsional strain. A balanced evaporation rate in the 10T:10B formulation supports sustained conductive pathways before mechanical fatigue becomes dominant at higher cycle counts. This [29] interprets the ageing trends discussed in Section 3.2, where differences in solvent architecture influence the stability of the mechanically conditioned conductive network under ambient exposure.

3.2. Ageing Behaviour of GNP/Ag Conductive Ink

This section examines the ageing behaviour of hybrid GNP/Ag conductive inks with different terpinol-to-butanol solvent ratios: 5T:10B, 10T:10B, and 15T:10B at room temperature. Resistance and resistivity trends were analysed to assess electrical stability after cyclic torsion.

3.2.1. Ageing Analysis for 5T:10B

Figure 6 presents the ageing behaviour of the 5T:10B formulation following cyclic torsional loading. Over the five-week ambient ageing period, resistance increased slightly from 0.46 Ω to 0.47 Ω, while resistivity rose from 2.73×10^{-5} Ω.m to 2.80×10^{-5} Ω.m. The overall resistivity variation remained within a narrow range of approximately 2–3%, indicating that the conductive network retained electrical continuity under room-temperature ageing conditions.

The limited magnitude of change suggests that ageing proceeded primarily through minor adjustment of conductive contacts rather than progressive electrical degradation. During the initial ageing stage, resistance and resistivity showed minimal variation, reflecting a network that had already been mechanically conditioned by prior cyclic torsion. Since the conductive ink had experienced junction rearrangement and partial contact modification during torsional loading, subsequent ageing occurred within an already adapted conductive network. Under these conditions, environmental exposure introduced only a limited additional electrical variation, after which the electrical response stabilised.

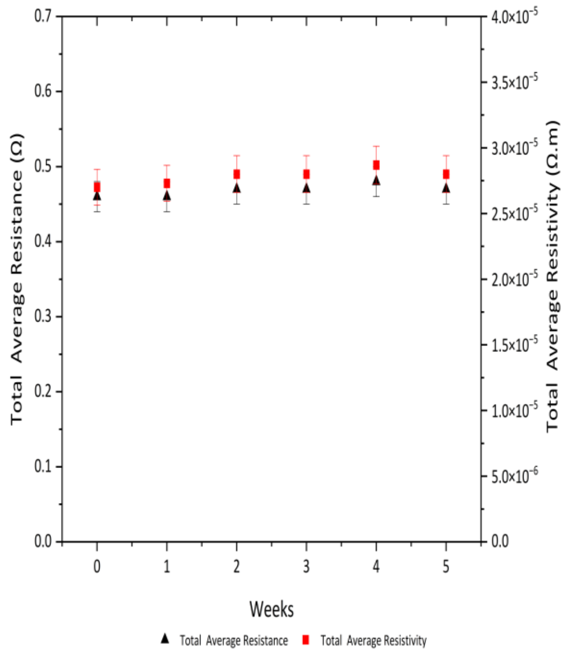

Figure 6. Total average resistance and resistivity for ageing sample 5T:10B in a five-week observation period. A ±5% error bar is embedded and illustrated across measured points.

In weeks two and three, resistance and resistivity approached a quasi-steady state, indicating that the conductive network had adapted to the static ambient environment. This behaviour is consistent with local contact-level rearrangement within the hybrid conductive network, where small changes in junction resistance were redistributed rather than accumulating at isolated conductive pathways. Such redistribution under static ageing conditions has been reported in particulate conductive systems and does not imply the formation of new defects or significant disruption of the conductive structure [30].

SEM observations of the 5T:10B formulation, shown in Figure 8, provide qualitative support for this interpretation. The micrograph reveals the presence of localised agglomerated regions and voids within the printed conductive layer, indicating that the conductive network is locally disrupted rather than uniformly compact. These features are consistent with the higher butanol content in the 5T:10B formulation, where faster solvent evaporation during curing can lead to local packing heterogeneity and reduced contact effectiveness between neighbouring conductive fillers. Such local disruptions increase junction resistance and contribute to the modest rise in resistance and resistivity observed during the early ageing stage.

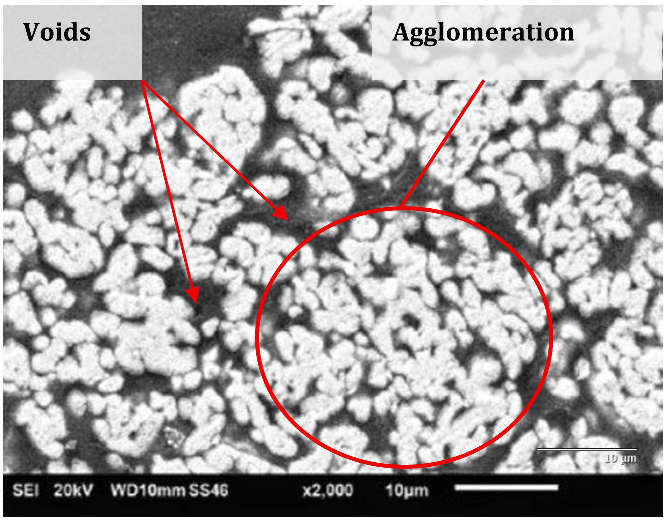

Figure 7. SEM micrograph of the 5T:10B hybrid GNP/Ag conductive ink after the week 5 ageing period

Under ambient ageing conditions, exposure to oxygen may also result in the formation of thin surface oxide layers on Ag particles and GNP. This oxidation contributes to incremental increases in contact resistance by modifying conductive interfaces. However, oxidation of the Ag at room temperature is widely reported to be self-limiting, producing thin surface layers rather than thick insulating phases that would severely disrupt charge transport [31]. Consequently, although oxidation occurs, its effect does not lead to drastic degradation of the conductive network. The presence of the GNP further mitigates the electrical impact of local contact degradation by providing alternative conductive pathways within the hybrid structure.

Overall, the 5T:10B formulation exhibits controlled ageing behaviour characterised by limited and non-progressive changes in resistance and resistivity. The combined effects of solvent-controlled film formation, mechanically conditioned conductive contacts, localised voids and agglomeration, and the oxidation-resistant nature of the hybrid GNP/Ag conductive ink network govern the observed electrical stability under ambient ageing conditions.

3.2.2. Ageing Analysis for 10T:10B

The resistance and resistivity trends for the 10T:10B composition during five weeks of ambient ageing following cyclic torsional loading are presented in Figure 8. The initial resistance and resistivity values were 0.37 Ω and 2.20x10^{-5} Ω.m, respectively. Both parameters remained largely unchanged up to week two, indicating limited sensitivity to ambient exposure during the early ageing stage. Between weeks two and three, resistivity increased slightly to 2.27x10^{-5} Ω.m, corresponding to a modest change of approximately 3%, while resistance increased to 0.38 Ω. This variation reflects a minor adjustment of conductive contacts rather than the onset of progressive degradation.

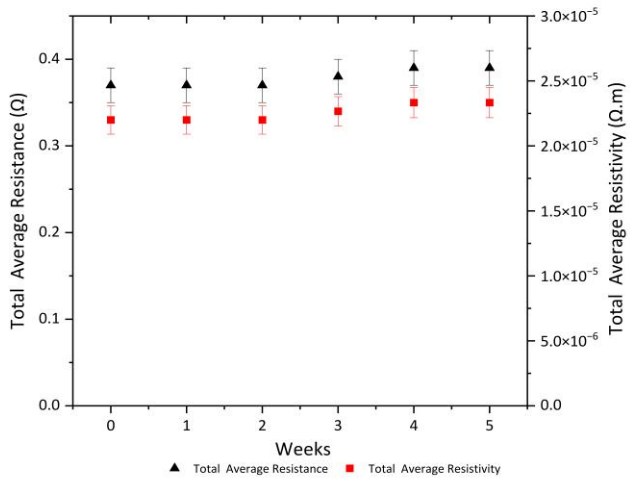

Figure 8. Evolution of total average resistance and resistivity of the 10T:10B formulation during a five-week ageing period. Error bars represent ±5% variation across the measured points.

The resistivity increased further to 2.33x10^{-5} Ω.m by week four, after which no additional increase was observed through week five. The total resistivity variation over the ageing period remained within a narrow range of approximately 5%. The stabilised response from week three onwards indicates that the conductive network had reached a steady electrical state under room-temperature ageing conditions. This behaviour distinguishes the 10T:10B formulation from compositions that exhibit continued drift or delayed degradation.

The stabilisation observed during ageing is closely associated with the solvent architecture of the 10T:10B formulation. The balanced terpinol to butanol ratio supports more uniform dispersion of conductive fillers during curing, which limits spatial variation in contact effectiveness across the printed layer. A more homogeneous particle distribution reduces the localisation of electrical sensitivity during ageing. Minor changes in individual contact resistance are distributed more evenly across the conductive network. This prevents these changes from accumulating at isolated junctions.

The SEM observations of the 10T:10B formulation, shown in Figure 9, provide qualitative support for this interpretation. The micrograph shows a relatively uniform distribution of conductive fillers with limited void formation. Although voids are visible, they are localised and do not form a continuous separation within the conductive layer. Such voids are known to increase resistance by interrupting conductive pathways [29]. However, in this case, their limited extent suggests that they do not dominate electrical transport. The presence of these voids is more plausibly associated with prior cyclic torsional loading rather than ageing itself, and their impact is mitigated by the overall continuity of the conductive network.

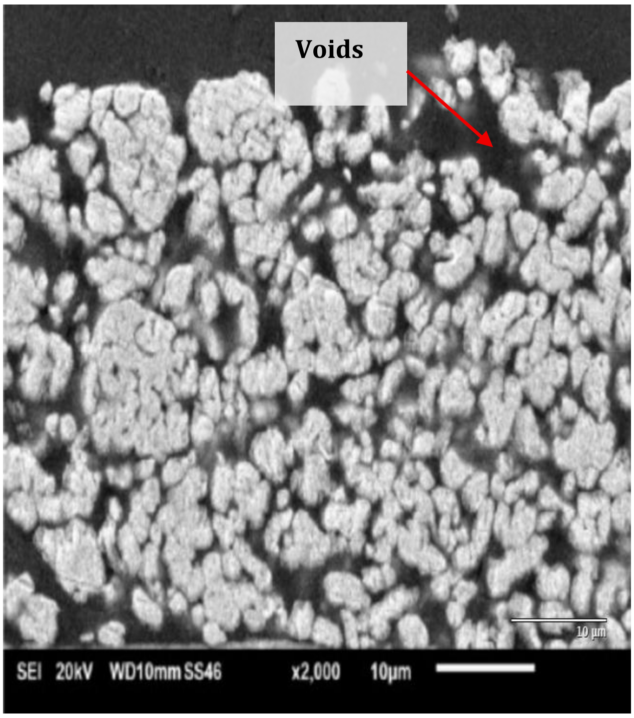

Figure 9. SEM micrograph of the 10T:10B hybrid GNP/Ag conductive ink after the week 5 ageing period.

The presence of the GNP plays an additional role in moderating ageing behaviour. The GNP are hydrophobic, whereas the Ag particles are not. This difference reduces the susceptibility of the conductive network to water uptake during ambient ageing. In the 10T:10B formulation, the combination of uniform particle dispersion and hydrophobic GNP limits moisture penetration into the conductive layer. As a result, environmental exposure introduces only limited perturbation at conductive contacts rather than widespread degradation.

Electrical continuity is further supported by the availability of parallel conduction pathways provided by the hybrid GNP and Ag network. Charge transport is not governed solely by individual Ag to Ag junctions. Instead, current redistribution through graphene pathways reduces the amplification of local contact variations. Consequently, resistance and resistivity exhibit only minor changes during the early ageing stage, followed by a stable response at later stages.

Overall, the ageing behaviour of the 10T:10B formulation is characterised by early stabilisation and limited electrical drift. The durability of electrical performance under room-temperature ageing is influenced by the formation of the film, which is governed by the solvent composition. This durability is further supported by the uniform dispersion of fillers and the hydrophobic properties of graphene nanoplatelets. The presence of multiple conductive pathways within the hybrid network also contributes to maintaining stable electrical behaviour.

3.2.3. Ageing Analysis for 15T:10B

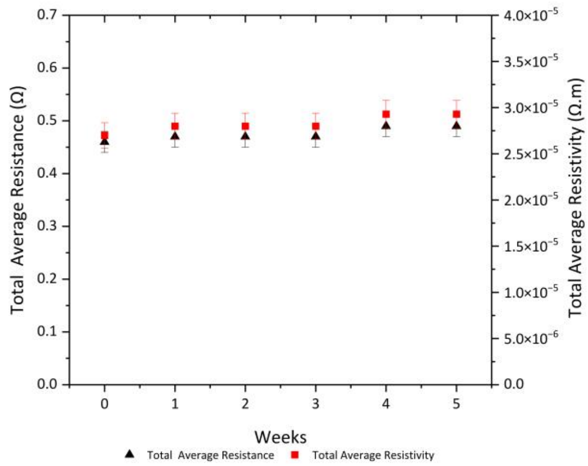

Figure 10. Total average resistance and resistivity graph for ageing sample 15T:10B.

Figure 11 presents the ageing behaviour of the 15T:10B formulation following cyclic torsional loading. Initial resistance and resistivity values of 0.46 Ω and 2.70×10^{-5} Ω.m increased to 0.49 Ω and 2.93×10^{-5} Ω.m after five weeks of room temperature ageing, corresponding to an overall change of approximately 9%. This limited magnitude of change indicates that the conductive network remained electrically continuous throughout the ageing period.

The ageing response is characterised by an early adjustment stage followed by stabilisation, rather than progressive electrical degradation. Changes in resistance and resistivity occurred mainly during the initial ageing phase, after which both parameters approached a steady trend within the measurement variation. This behaviour suggests that ageing primarily involves minor redistribution of existing conductive contacts instead of the formation of new degradation pathways.

Compared with the 5T:10B and 10T:10B formulations, the 15T:10B sample exhibited a slightly larger resistivity increase during ageing. This difference is consistent with variations in solvent composition influencing the final contact configuration established during curing. Differences in ageing response for the 15T:10B formulation can be associated with the higher terpinol content, which influences how the cured conductive network responds to ambient exposure.

Overall, the 15T:10B formulation demonstrates controlled ageing behaviour, with resistivity changes remaining limited and non-progressive over time. The observed trend confirms that environmental exposure at room temperature induces only modest electrical variation within the hybrid GNP/Ag conductive ink network.

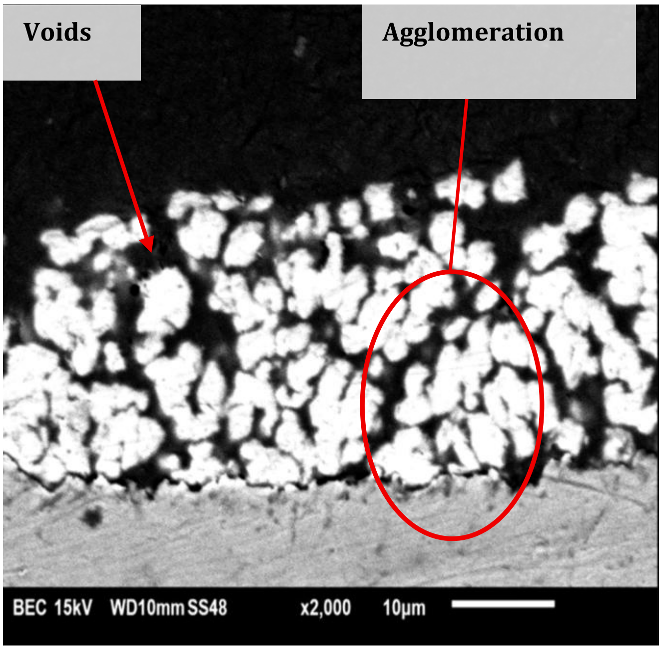

Figure 11. SEM micrograph of the 10T:10B hybrid GNP/Ag conductive ink after the week 5 ageing period

4. CONCLUSION

This study examined the influence of the terpinol to butanol solvent ratio on the electrical behaviour of hybrid GNP/Ag conductive ink under cyclic torsional loading and subsequent ageing. Across all formulations, resistance increased with increasing torsional cycles. Resistivity also increased as the number of torsional cycles increased. These trends reflect the progressive modification of conductive contacts within the printed ink during mechanical deformation. The magnitude and rate of this increase were strongly dependent on the solvent composition.

Following cyclic torsion, only minor changes in resistivity were observed during room-temperature ageing. After the initial ageing stage, the electrical response stabilised. No further increase in resistivity was detected, and cumulative degradation was observed during room-temperature ageing. This outcome indicates that the conductive pathways established after mechanical loading remained largely preserved under ambient environmental exposure.

Among the formulations studied, the 10T:10B solvent ratio provided the most favourable balance between dispersion uniformity, electrical performance, and mechanical durability. The balanced solvent composition supported a conductive network that was more resilient to torsional cycling while maintaining stable electrical behaviour during ageing. These findings demonstrate that optimisation of the terpinol to butanol ratio is a key parameter for achieving durable and reliable hybrid GNP/Ag conductive inks for flexible electronic applications.

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