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Relationship between voltage and resistance in hybrid nano-conductive ink on different substrates in wet and dry conditions

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ABSTRACT

Hybrid graphene nanoplatelet/silver (GNP/Ag/SA) conductive inks are increasingly used in flexible electronics, yet there is limited understanding of how substrate type, solvent composition, and moisture exposure jointly control the electrical performance on metal and polymer substrates. This work aims to clarify how terpinol content (5T, 10T, 15T) and substrate properties of copper (Cu), polyethylene terephthalate (PET), and thermoplastic polyurethane (TPU) influence voltage, resistance, and resistivity of screen-printed GNP/Ag/SA tracks under dry and post-immersion wet conditions. GNP/Ag/SA inks were formulated with fixed butanol and varied terpinol contents, printed on Cu, PET, and TPU, and characterized using electrical measurements, adhesion evaluation, and microstructural observations to relate resistivity trends to morphology, surface energy, and hygroscopic behavior. The Cu substrate showed the best performance, with Cu 10T achieving the lowest dry resistivity of approximately 1.2×10-5 Ω.m and Cu 15T the lowest wet resistivity of approximately 2.0×10-5 Ω.m, supported by dense, well-adhered microstructures. The PET exhibited higher resistivity values up to about 10-3 Ω.m and clear degradation after water immersion, while TPU showed very high or unmeasurable resistivity in wet conditions caused by severe ink loss and hygroscopic swelling, highlighting the important role of substrate surface energy and moisture response in determining the reliability of GNP/Ag/SA inks for applications in humid or wet environments.

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1. INTRODUCTION

Conductive ink is typically created by infusing traditional ink with conductive materials like graphite, silver flakes, or nanoparticles into a liquid vehicle, enabling printed circuits and components on a wide range of substrates without conventional wiring [1]. Carbon-based inks, especially those using graphene nanoplatelets (GNP), have attracted considerable attention because they offer a promising alternative to conventional metal-based inks by combining good conductivity with flexibility, chemical stability, and potential cost advantages [2], [3]. Since the isolation of graphene in 2004 by Geim and Novoselov, GNP and related graphene family materials such as graphene oxide, reduced graphene oxide and few-layer graphene have been widely explored for printed and flexible electronics for their high intrinsic electrical conductivity, large specific surface area, and excellent mechanical properties [4]-[7]. Despite of GNP's advantages, investigation findings by Naghdi *et al.* [8] mentioned achieving uniform dispersion of the fillers is the most challenge part including melt blending, solution mixing, in situ and latex blending methods and Vashist *et al.* [3] reported GNP also comes with issues such as stability, dispersion in water and annealing temperature.

Hybridization of GNP with highly conductive metals, particularly silver (Ag), has therefore appeared as an effective strategy to enhance electrical performance while retaining some of graphene's mechanical robustness and flexibility [9], [10]. The silver acetate (SA) is an attractive additive in such hybrid systems because it acts as a metal precursor that decomposes during curing to generate fresh Ag, promoting sintering and neck growth between Ag particles, filling gaps in the GNP/Ag network, and improving particle–particle and ink–substrate contact as this enables tuning of microstructure and connectivity at relatively low curing temperatures, which is important for flexible polymer substrates with limited thermal tolerance [9]. In GNP/Ag/SA hybrid inks, 1-butanol and terpinol are commonly employed as organic solvents because they dissolve silver compounds, aid dispersion of GNP and Ag flakes, and tune viscosity and drying behaviour for screen or paste printing. Butanol tends to promote more uniform particle dispersion and lower resistivity when optimally balanced, whereas terpinol, with higher viscosity and different evaporation behaviour, improves printability and acts as a binding medium but can hinder dispersion and increase agglomeration if present in excess, thereby strongly influencing the final microstructure and electrical performance [10]. Recent work by Ismail *et al.* [11] and Noor *et al.* [12] has shown that GNP/Ag and GNP/Ag/SA hybrids can achieve significantly higher conductivity than pure GNP inks, although their resistance still increases under cyclic torsion and bending, indicating that mechanical integrity and interfacial bonding remain critical design constraints. Ismail *et al.* [11] also reported that screen-printed GNP/Ag inks on copper (Cu), cured at 250 °C for 1 hour, exhibited rising resistance under repeated torsional stress, while Noor *et al.* [12], [13] linked resistance and resistivity increases during cyclic bending to microstructural damage and breakdown of bonding within the GNP-based network [11]-[13]. The GNP hybrid inks are typically multi-component systems comprising GNP, Ag, and organic solvents such as butanol and terpinol, and optimization of composition is important to achieve improved adhesion and electrical conductivity. For example, Hussin *et al.* [14] reported a substantial reduction in resistivity when using smaller Ag particle sizes (5 µm) compared with larger particles (25 µm), highlighting the influence of particle size on electrical performance [14]. Under dry conditions, GNP/Ag hybrid inks generally show stable electrical properties, but under cyclic mechanical loading conductivity ultimately degrades due to microstructural changes. Moreover, Noor *et al.* [15] indicated that humidity can affect the structural integrity and adhesion of conductive paths, leading to increased resistance. However, detailed studies of GNP/Ag/SA hybrids on different substrates under wet conditions remain scarce.

The existing literature has predominantly focused on conductivity improvements in dry conditions which systematic studies that link SA assisted hybridization to adhesion, morphology, and stability under moisture exposure across different substrate types such as metal versus low surface energy versus hygroscopic polymers are still lacking. In particular, the combined influence of substrate surface energy and hygroscopicity behaviour especially in TPU, curing constraints imposed by substrate thermal stability, and solvent composition such butanol/terpinol ratio on the wet and dry electrical performance of GNP/Ag/SA hybrid inks has not been comprehensively addressed.

This work addresses a clear gap in understanding how substrate type Cu, polyethylene terephthalate (PET), thermoplastic polyurethane (TPU), surface energy, curing conditions, moisture exposure, and terpinol content are 5T, 10T, 15T collectively influence the morphology, adhesion, and electrical stability of GNP/Ag hybrid conductive inks, particularly under wet conditions. The practical problem is that printed conductive tracks often degrade after moisture exposure, especially on low surface energy and hygroscopic polymers like PET and TPU, leading to poor adhesion, discontinuous networks, unstable or unmeasurable resistivity, and increased voltage drop, with limited guidance available for material and process selection. This study aims to evaluate and compare GNP/Ag hybrid inks with different terpinol contents on Cu, PET, and TPU by measuring voltage, resistance, and resistivity in dry and wet conditions, assessing adhesion and morphology, and relating these to surface energy, curing behaviour, and hygroscopic effects, in order to identify suitable substrate–ink combinations for applications where moisture and substrate limitations are needed.

2. METHOD

2.1. Conductive ink preparation

Initially, conductive ink must be prepared in a powdered state prior to the three formulations of terpinol and 1-butanol ratio mixtures. The powder preparation process is designed to achieve uniform dispersion of conductive materials while maintaining optimal particle size distribution for printing. The hybrid GNP/Ag powder and conductive ink were prepared according to previous study conducted by

Jori *et al.* [16]. The materials used to produce GNP/Ag hybrid powder and conductive ink were shown in Table 1.

Table 1. Materials used in formulation of GNP/Ag hybrid powder and conductive ink

Materials	Details
Graphene (GNP)	25 µm particles size with the surface area of 50 to 110 m^2/g
Silver (Ag)	10 µm particles size, 99.9% trace metals basis, 1.59 µΩ/cm, 20 °C resistivity
Silver acetate (SA)	<10 µm particles size, 99.99% trace metal basis acid silver salt, 4.2-51 µΩ/cm resistivity
Ethanol	Denatured 99%, 46.07 g/mol molecular mass
1-Butanol	99.9% butyl alcohol, 74.12 g/mol molecular mass
Terpinol	Pine oil contains 65% α, 10% β, 20% γ, 154.25 g/mol molecular mass

First, 0.010 g of GNP is measured and mixed with 10 ml of ethanol in a clean beaker, then covered with aluminum foil to prevent evaporation. The mixture undergoes ultrasonic mixing for 10 minutes. Next, 0.8584 g of silver flakes (SF) is added, and the mixture is sonicated for 1 hour for uniform distribution. Afterward, 0.084 g of SA is incorporated and treated ultrasonically for another hour at 200 rpm and 70 °C.

Following drying, the mixture is cured at 250 °C for 1 hour and then cooled before being pounded in a mortar to refine the powder for a smooth texture. The refined powder is weighed, and the container weight recorded before adding terpinol and butanol in precise amounts. The formulation is blended in a planetary centrifugal mixer at 2,000 rpm for 3 minutes to achieve a homogeneous paste. Finally, the container is labeled and sealed to prevent ink evaporation. Figure 1 illustrates process of powder and paste for GNP hybrid conductive ink.

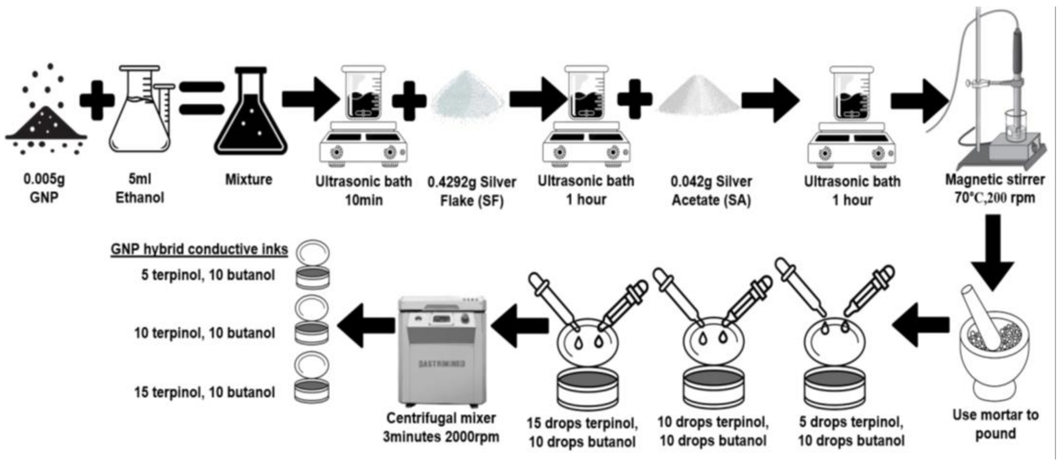


Figure 1. Conductive ink preparation process

In Table 2, there are three formulations were developed to study the electrical effect of terpinol-to-butanol ratio on ink properties. The baseline sample is 10T/10B and butanol ratio is fixed to 10 drops throughout this experiment. In a comparative study of solvent effects on GNP/Ag conductive inks printed on Cu substrates, Jori *et al.* [16] in 2024 reported that varying terpinol volume from 5 to 15 drops resulted in a pronounced reduction in resistivity relative to equivalent butanol variations. The formulation with 5 drops of terpinol exhibited the lowest resistivity, indicating enhanced electrical conductivity. The use of terpinol-based inks particularly showcased advantages in terms of viscosity, flowability, resolution, and surface morphology [16].

Table 2. The formulation of GNP/Ag hybrid conductive inks

Formulation	Terpinol (T)	Butanol (B)	Expectation
5T/10B	5 drops ~ 0.325 ml	10 drops ~ 0.65 ml	Lower viscosity for fine detail printing
10T/10B	10 drops ~ 0.65 ml	10 drops ~ 0.65 ml	Balanced viscosity as baseline sample
15T/10B	15 drops ~ 0.975 ml	10 drops ~ 0.65 ml	Higher viscosity for improved substrate adhesion

2.2. Sample preparation

Figure 2 illustrates the sample preparation has been done by 60 µm stencils screen printing method using three GNP hybrid conductive ink formulations on three different substrates. The Cu, PET, and TPU substrates were cut to 12 cm×1 cm and cleaned with isopropyl alcohol. It required the conductive ink to be applied on top of it, then to be pressed with consistent pressure to produce a square shape written with ink thickness of 60 µm on the substrates. The stencils are then carefully removed to prevent unwanted spreading before curing process.

Figure 3 shows conductive ink applies to substrates by using stencils printing on five samples. Once it has printed on the substrates, labelling as in Table 3 and cure process executed at 260 °C for 3 hours for the Cu substrate as study demonstrated by Noor *et al.* [17] shows curing GNP/Ag/SA inks at temperatures like 240-260 °C for durations of 4-6 hours showed effective curing and conductivity, although adhesion strength may decrease at higher temperatures, indicating a trade-off between conductivity and adhesion. Additionally, else at 100 °C for 1 hour for PET and TPU substrates to prevent them from substrate deformation and shorter time minimizes thermal stress occurrence [18]. The actual after cured sample are shown in Figures 4-6.

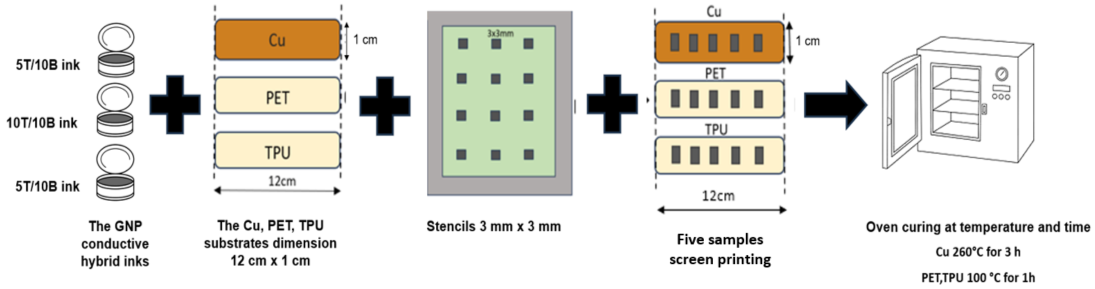


Figure 2. Sample preparation process

Figure 3. Stencils printing process on substrate

Table 3. The formulation of GNP/Ag hybrid conductive ink

Formulation/Substrates	Cu	PET	TPU
5T/10B	Run 1	Run 4	Run 7
10T/10B	Run 2	Run 5	Run 8
15T/10B	Run 3	Run 6	Run 9

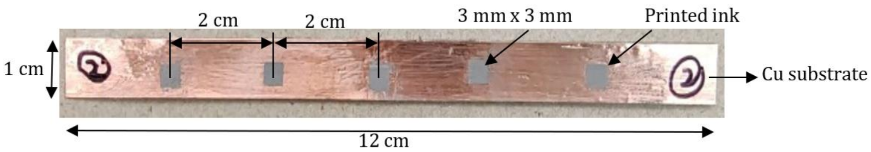


Figure 4. Five points of printed ink on Cu substrate

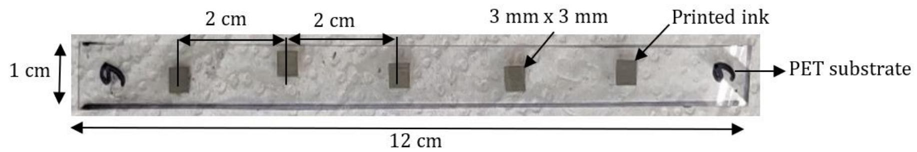


Figure 5. Five points of printed ink on PET substrate

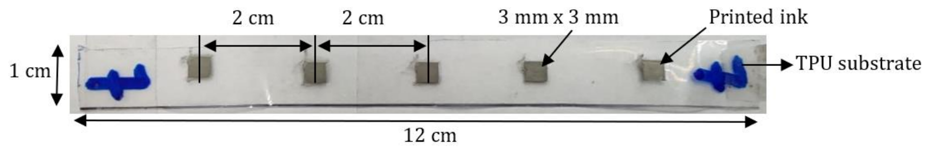


Figure 6. Five points of printed ink on TPU substrate

2.3. Characterization method

2.3.1. Voltage drop measurement

Basically, in an electrical and electronic circuit, current flows through when power supplied across and energy dissipated as heat, then voltage drop happened because of the existence of resistance or impedance components or conductors. This explained by Priyadarsini *et al.* [5] and Olabi *et al.* [6].

The equipment required for this measurement includes a two-point probe with a minimum resolution of 0.1 mV, a regulated DC power supply with a 0-30 V range and datasheet. After cured sample will be waited cold prior this measurement. The samples will be measured in two conditions, dry and wet condition. Figure 7 demonstrates voltage measurement at five points in the dry condition, which all samples will be measured point-to-point within 1-V to 9-V voltage supply connected end-to-end of substrate and by using digital multimeter with a minimum resolution of 0.1 mV.

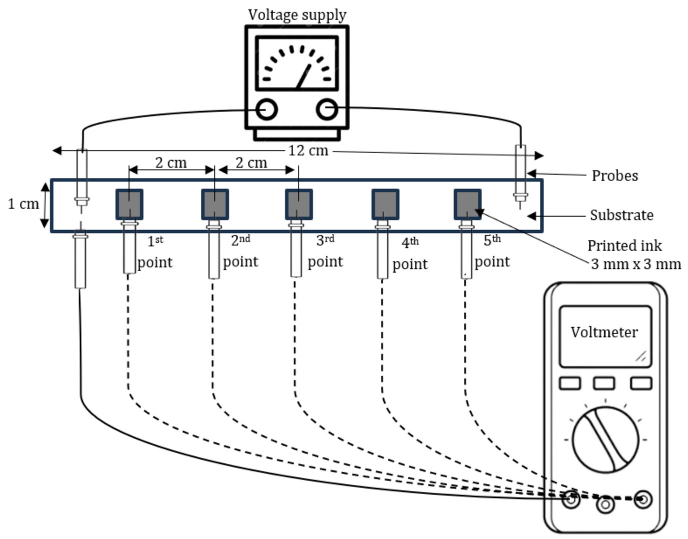


Figure 7. Voltage measurement on a substrate at five points in dry condition

Figure 8 illustrates the experimental procedure and voltage measurement setup used under wet conditions. After the dry condition is completed, the samples are exposed to tap water for five minutes and then dried for another five minutes according to ASTM D870, as shown in Figure 8(a) and Figure 8(b). The voltage measurements are then repeated within a 1-V to 9-V voltage supply using a digital multimeter, as illustrated in Figure 8(c). This procedure ensures consistent results across varying conditions and enables accurate comparison of the samples' conductivity.

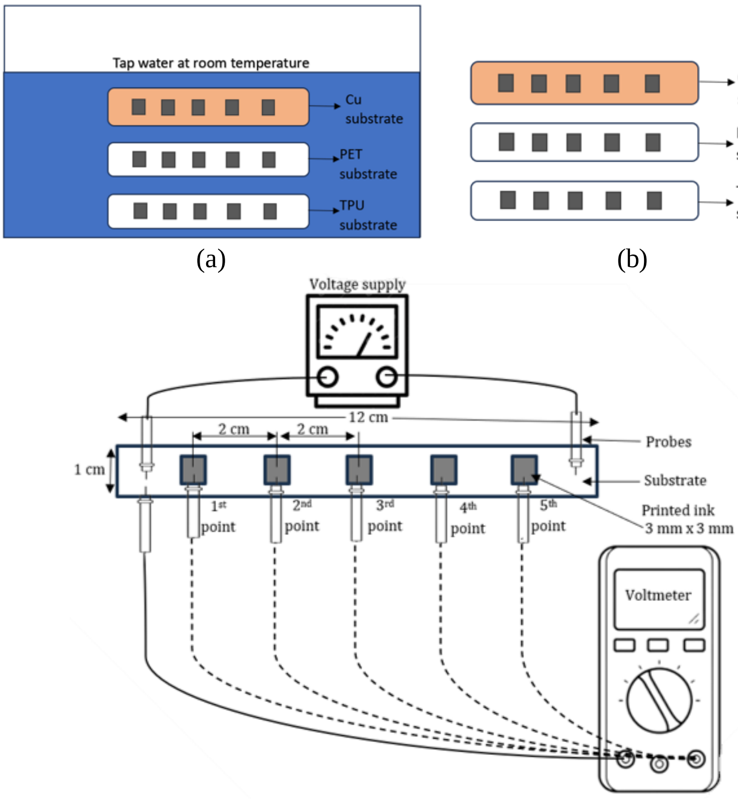


Figure 8. Voltage measurement on a substrate at five points in wet condition; (a) soaked for five minutes, (b) dried for five minutes, and (c) voltage measurement by using digital multimeter

2.3.2. Resistant measurement

The most widely used techniques for measuring the electrical resistance of GNP ink are the two-point probe method as mentioned by Cultrera *et al.* [19]. There are two conditions involved in the measurement, dry and wet conditions. The procedure of resistance measurement in dry condition is shown in Figure 9. Both probes will be placed on two points and connected to a digital multimeter in the ohmmeter range. The reading will be collected at five points and recorded in the datasheet. In the wet condition, all samples will be soaked for five minutes in the tap water before let dried for another five minutes. This measurement will be repeated and the data will be used in the calculation to obtain resistivity value.

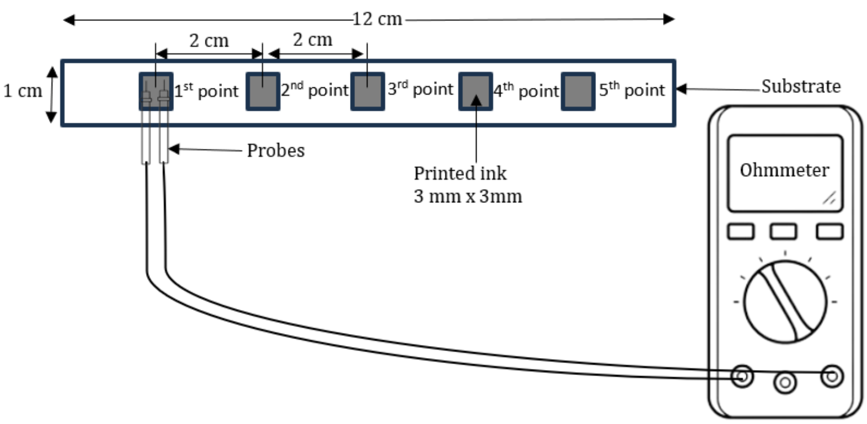


Figure 9. Resistance measurement on a substrate at five points in dry condition

2.3.3. Resistivity calculation

In research paper written by Ghorbani and Taherian [20], resistivity, denoted by the symbol ρ (rho), is a fundamental property of materials that describes ability to resist the flow of electrical current as in Figure 10. Similarly, Ismail *et al.* [11] has investigated the resistivity of GNP/Ag hybridization conductive ink under bending stress, which can also provide insights into the relationship between adhesion and resistivity. This property is inversely related to electrical conductivity, which measures a material's capacity to allow the flow of electric charge. Material's resistivity is influenced by the geometric dimensions, as expressed by the formula $p = \frac{RA}{l}$, where R is the electrical resistance, A is the cross-sectional area, and ℓ is the length of the material.

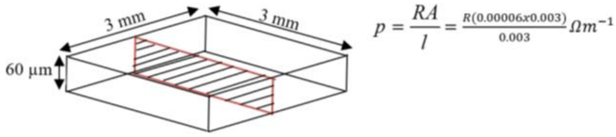


Figure 10. Resistivity calculation on a square shape ink

3. RESULTS AND DISCUSSION

3.1. Average results of measurements

Table 4 presents average data of voltage, resistance, and resistivity for the various substrates under dry and wet conditions. This information is important in assessing the performance of various materials in practical applications, where moisture can have a profound impact on their electrical properties. The results account for trends in the electrical behaviour of each substrate and formulation thereby simplifying the selection of suitable materials according to specific application demands.

Table 4. Average voltage, resistance and resistivity for different substrates in dry and wet conditions

RUN	DRY voltage	WET voltage	DRY resistance	WET resistance	DRY resistivity	WET resistivity
5T Cu	0.0020	0.0031	0.2200	0.6000	1.32×10^{-5}	3.60×10^{-5}
10T Cu	0.0022	0.0036	0.2000	0.4000	1.20×10^{-5}	2.40×10^{-5}
15T Cu	0.0024	0.0033	0.2600	0.3400	1.56×10^{-5}	2.04×10^{-5}
5T PET	0.0021	0.0029	1.4600	4.4000	8.76×10^{-5}	2.64×10^{-4}
10T PET	0.0017	0.0015	4.1400	5.2500	2.48×10^{-4}	3.15×10^{-4}
15T PET	0.0030	0.0025	1.9600	0.9000	1.18×10^{-4}	5.40×10^{-5}
5T TPU	0.0024	0.0030	12.6000	0.7500	7.56×10^{-4}	4.50×10^{-5}
10T TPU	0.0017	0.0023	116.6400	Unable to measure	7.00×10^{-3}	Unable to measure
15T TPU	0.0016	0.0021	Unable to measure	Unable to measure	Unable to measure	Unable to measure

3.2. Analysis of substrate effects on electrical conductivity across 5T, 10T, and 15T

Figures 11-13 illustrated data obtained from Table 4 to investigates the linking between substrates and electrical performance for each formualtion of 5T, 10T, and 15T. Figure 11 shows the electrical performance for 5T formulation under dry and wet across substrates. Across substrates, voltage drop and resistivity exhibit an upward trend from dry to wet conditions, with Cu demonstrating the best performance with 1.32×10^{-5} Ω.m - 3.60×10^{-5} Ω.m. Figure 12 presents the electrical performance of the 10T formulation under both conditions. The Cu substrate maintained good conductivity, with resistivity values ranging from 1.20×10^{-5} Ω.m to 2.40×10^{-5} Ω.m. In contrast, the PET substrate exhibited acceptable resistivity between 2.48×10^{-4} Ω.m and 3.15×10^{-4} Ω.m, whereas the TPU substrate showed unstable resistivity across some samples and became unmeasurable after water immersion. Figure 13 shows that the TPU substrate experienced the most severe degradation, with none of the samples measurable for either resistivity or voltage drop. The Cu substrate remained the best performer, exhibiting low resistivity values between 1.56×10^{-5} Ω.m and 2.04×10^{-5} Ω.m, while the PET substrate displayed the highest resistivity compared to its 5T and 10T counterparts.

The Cu generally exhibits a relatively high surface energy, which promotes good wetting and strong interfacial adhesion of conductive inks on its surface, enabling the formation of continuous, well-bonded metallic tracks with low resistivity for printed and flexible electronics [21]. Figure 14 presents a schematic illustration of the effect of substrate surface energy on the interfacial contact area and adhesion behavior of GNP/Ag conductive inks. As for this, Cu substrate promotes strong wetting and adhesion of the GNP/Ag conductive ink, enabling a continuous and mechanically robust conductive network as in Figure 14(a).

This intimate interfacial contact reduces interfacial contact resistance, thereby lowering the overall resistivity and minimizing voltage drop along the printed tracks.

Adhesion quality also not only depends on substrate's surface energy, but also depends on the curing temperature and duration as well which together control the formation and stability of the conductive network. Curing conditions strongly influence adhesion because solvent evaporation, particle sintering, and binder crosslinking occur during thermal treatment. Suboptimal curing can lead to residual solvent, microcracking, or a weakly bonded interface, whereas appropriate curing promotes densification of the GNP/Ag network and stronger interfacial bonding, improving both mechanical integrity and electrical conductivity [17].

The Cu exhibits good thermal stability, which allows curing at 260 °C for 3 hours without degrading the substrate and promotes stronger interfacial bonding between the GNP/Ag ink and the Cu surface. The resulting strong adhesion helps maintain the ink thickness even after water immersion, as evidenced by the scanning electron microscopy (SEM) observations.

However, PET is a low surface energy substrate and TPU exhibits even lower surface energy, leads to smaller contact area interfacial bond which reduces ink–substrate adhesion and leads to higher resistivity compared to Cu as shows in Figure 14(b). In addition, their thermal sensitivity limits the curing conditions to around for 100 °C 1 hour, further constraining interfacial bonding and the development of a well connected conductive network.

These combined factors result in lower ink performance across the formulations, as evidenced by degradation observed in some or all samples after water immersion as in Figure 15. Although Wang *et al.* [22] identified distinct phases in moisture adsorption for uncoated PET film, indicating its stability during moisture exposure. However, Watanabe *et al.* [23] stated that water acts as a conductor, increasing the electrical conductivity of PET. Water may increase dielectric constant of PET and change the electrical behavior of conductive layers on its surface which potentially leading to voltage fluctuations. Though, this effect is generally minimal because of PET's low water absorption rate which around 0.5%.

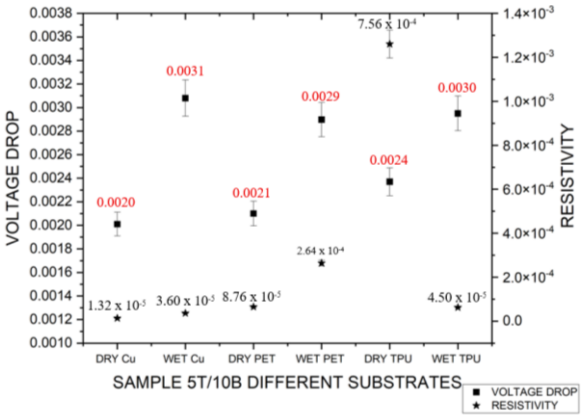


Figure 11. Voltage drops vs resistivity on different substrates on 5T/10B formulation

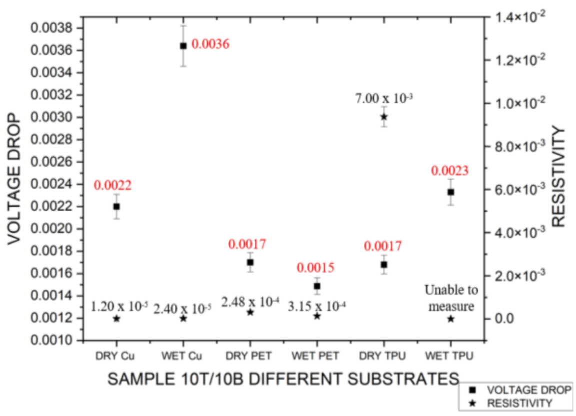


Figure 12. Voltage drops vs resistivity on different substrates on 10T/10B formulation

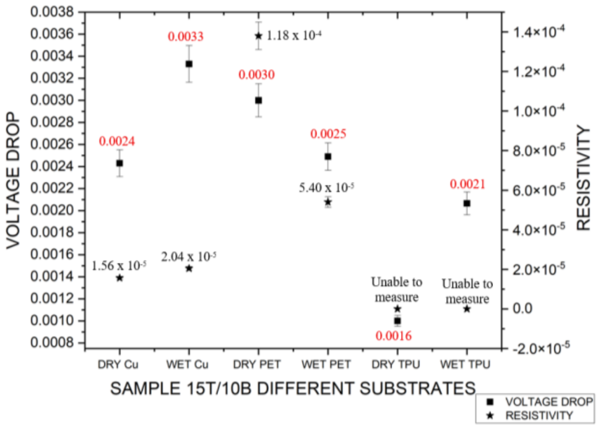


Figure 13. Voltage drops vs resistivity on different substrates on 15T/10B formulation

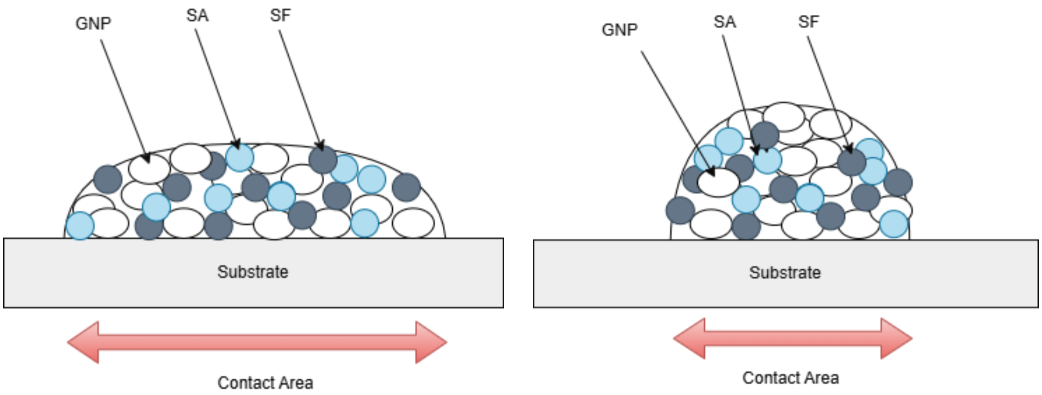


Figure 14. Surface energy illustration for GNP/Ag ink (a) high surface energy substrate produce larger contact area and better adhesion and (b) low surface energy substrate produce smaller contact area and poorer adhesion

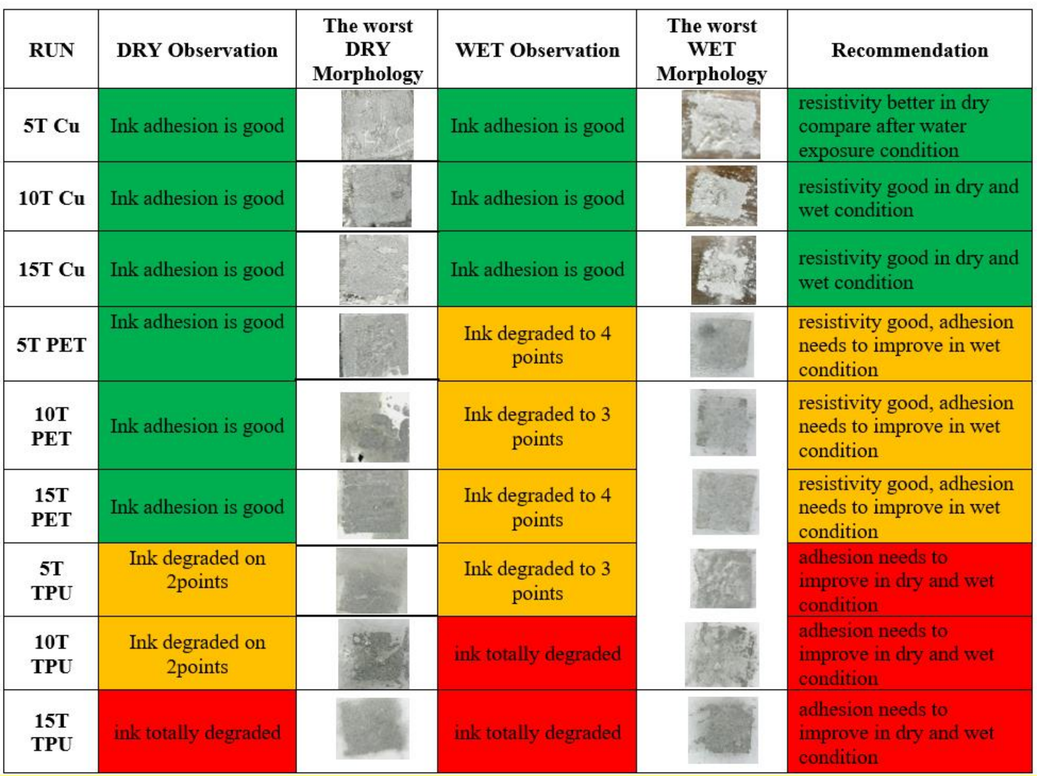
3.3. Adhesion assessment under dry and wet conditions

The Figure 15 is consolidates the data to describe the relative of adhesion each formulation on various substrates under dry and wet conditions. Based on this Figure, there are three groups classified there are good adhesion under dry and wet conditions, good adhesion under but degraded under wet conditions, and degraded under both dry and wet conditions.

Performance of GNP conductive ink relies on several adhesion mechanisms to ensure strong bonding between the ink and the substrate in maintaining the electrical properties of the ink under various conditions. A good adhesion will create a stable conductive pathway between conductive ink and substrate, thus maintaining low resistance and consistence voltage across the ink layer.

Conductive pathway in GNP fulfills the percolation theory as it describes the conductive particles must reach percolation threshold to form continuous pathways for electron flows and reducing electrical resistance as mentioned by Saad *et al.* [24]. It also protects the ink from moisture changes, where water may disrupt conductive pathway if the ink adhesion is weak then may lead increase voltage drop and resistance.

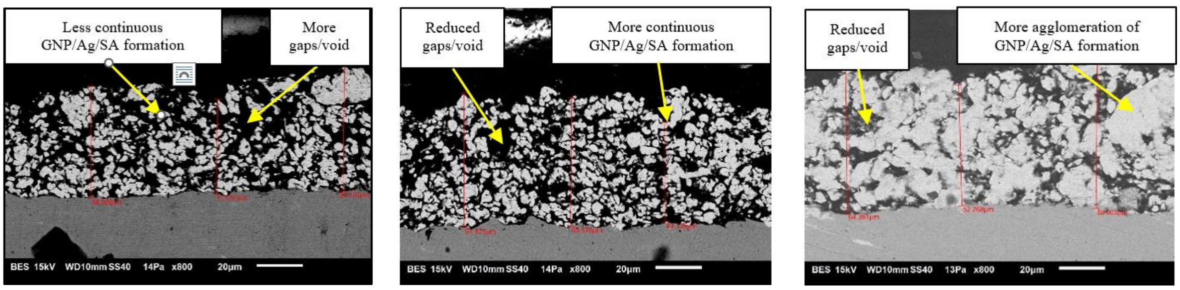
Conductive inks, particularly those based on GNP, rely strongly on solvents for optimal performance. Noor *et al.* [12] say terpinol is an effective solvent that facilitates the dissolution and dispersion of conductive materials, such as Ag particles. The solvent enhances the fluidity of the ink, allowing it to be easily deposited on various substrates. In addition, terpinol stabilizes the ink and also possesses a lower toxicity profile compared to other solvents. This results in a homogeneous and stable conductive ink, and this is what is required for quality printing and consistent electrical performance. Regarding the viscosity of the ink, research by Jori *et al.* [16] indicates that higher concentrations of terpinol can render ink viscosity greater, which is ideal for some forms of printing while potentially rendering others challenging. Hence, the balance between terpinol and butanol must be optimized for achieving printability, substrate adherence, and overall conductivity. Sanchez-Duenas *et al.* [25] emphasized that the compatibility of all components in the formulation of the ink is key in ensuring the maintenance of desired properties, which impacts the electrical property of the final product.



RUN	DRY Observation	The worst DRY Morphology	WET Observation	The worst WET Morphology	Recommendation
5T Cu	Ink adhesion is good		Ink adhesion is good		resistivity better in dry compare after water exposure condition
10T Cu	Ink adhesion is good		Ink adhesion is good		resistivity good in dry and wet condition
15T Cu	Ink adhesion is good		Ink adhesion is good		resistivity good in dry and wet condition
5T PET	Ink adhesion is good		Ink degraded to 4 points		resistivity good, adhesion needs to improve in wet condition
10T PET	Ink adhesion is good		Ink degraded to 3 points		resistivity good, adhesion needs to improve in wet condition
15T PET	Ink adhesion is good		Ink degraded to 4 points		resistivity good, adhesion needs to improve in wet condition
5T TPU	Ink degraded on 2points		Ink degraded to 3 points		adhesion needs to improve in dry and wet condition
10T TPU	Ink degraded on 2points		ink totally degraded		adhesion needs to improve in dry and wet condition
15T TPU	ink totally degraded		ink totally degraded		adhesion needs to improve in dry and wet condition

Figure 15. Overall adhesion result comparison for different substrates in dry and wet conditions

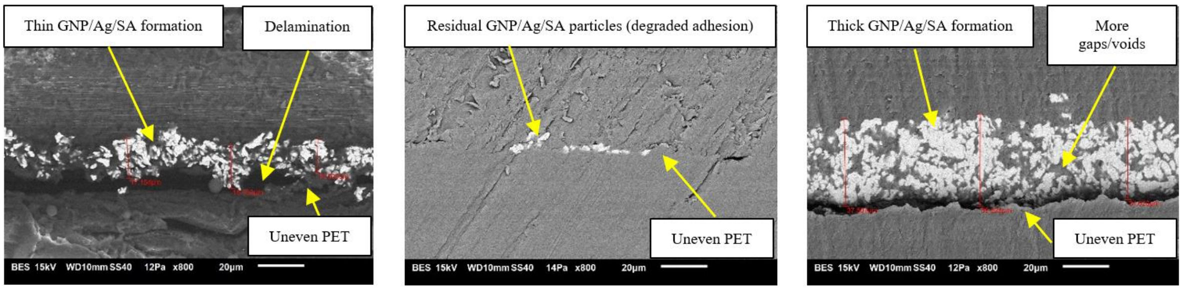
Figures 16-18 presents SEM images after water immersion used to evaluate the morphology across the different substrates. For the Cu substrate (Figure 16(a)–(c)), Figure 16(b) evidence the 10T formulation appears the most uniform, with particles in close contact and neither pronounced agglomeration nor large gaps/voids, in contrast to both 5T and 15T in Figure 16(a),(c). Consistent with these cross-sectional observations, the 5T formulation exhibited higher resistivity than the 15T ink. The 5T sample showed a more porous and non-uniform particle distribution with larger gaps in the conductive path, whereas the denser, though more agglomerated, 15T network provided a more continuous percolation pathway for electron transport. This evidence that higher terpinol 10T and 15T produce better GNP, Ag, and SA formation that enables continuous conductive pathways and better eletrical performance.



(a) (b) (c)

Figure 16. SEM results on Cu substrates (a) 5T, (b) 10T, and (c) 15T

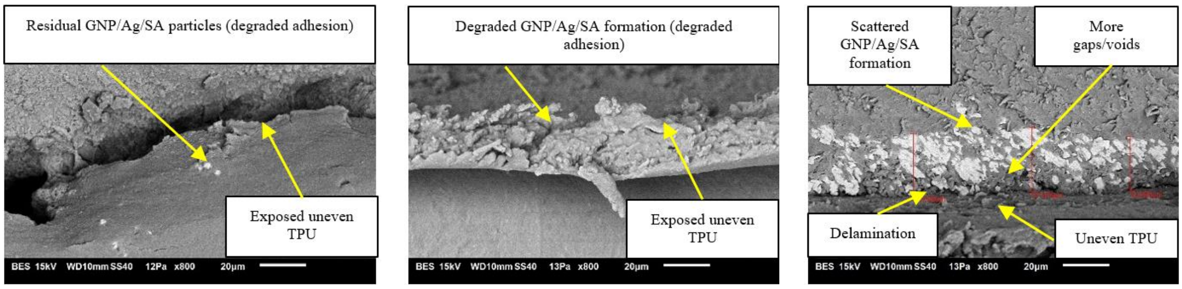
Figure 17 presents PET 5T, 10T, and 15T respectively. On PET Figure 17(a-c), all terpinol contents suffer from limited adhesion because of the low substrate surface energy and restricted curing at 100 °C 1 hour. Figures 17(a) shows the 5T ink forms a thin, highly discontinuous layer with large gaps, indicating poor wetting and weak interfacial bonding. The 10T formulation in Figure 17(b) shows only sparse, island-like coverage, suggesting even poorer wetting and very limited contact area. Although Figure 17(c) on 15T produces a thicker layer with better areal coverage, the structure is highly porous and rough, with many voids, so the conductive network remains discontinuous. These morphologies, combined with weak adhesion, explain the relatively high resistivity and pronounced degradation after water immersion for all PET samples.



(a) (b) (c)

Figure 17. SEM results on PET substrates; (a) 5T, (b) 10T, and (c) 15T

On TPU, all three terpinol formulations performed poorly because of TPU's very low surface energy and low curing temperature 100 °C 1 hour but in slightly different ways as in Figure 18. Figure 18(a) shows the 5T ink after water immersion leaves only a few residual particles on the TPU surface giving weak, easily broken percolation paths and unstable resistance. At 10T in Figure 18(b) on the other hand, the GNP/Ag layer is almost completely removed, exposing the TPU surface so unable to be established the electrical measurement. Figure 18(c) with 15T, the ink formed a scattered thicker layer than other formulations, but the structure remained porous, partially delaminated and irregular with weak interfacial bonding and discontinued conductive network causing unable to perform resistivity measurement.



(a) (b) (c)

Figure 18. SEM results on TPU substrates; (a) 5T, (b) 10T, and (c) 15T

These observations suggest that increasing terpinol content can enhance initial GNP/Ag/SA ink thickness and coverage on TPU, but it cannot overcome the fundamental limitations imposed by TPU's very low surface energy and hygroscopic swelling, so adhesion and electrical stability remain poor for all terpinol levels. Allami *et al.* [26] demonstrated that water ingress into TPU causes swelling and alters its internal structure, which in turn affects electrical and mechanical properties. Zweiri *et al.* [27] further reported that water immersion significantly modifies TPU properties, as water molecules disrupt intermolecular interactions between polymer chains, reducing Young's modulus and tensile strength.

These substrate level changes can worsen the impact of moisture on conductive inks applied to TPU as the TPU softens and swells, it becomes less dimensionally stable after immersion, which may promote cracking or delamination at the ink–substrate interface and disrupt the conductive path, thereby increasing voltage drop and resistivity across the printed layer [26], [27].

Figure 19 presents the adhesion behaviour of ink printed on the TPU substrate before and after moisture exposure. In the corresponding SEM images, the rough and irregular layer observed beneath the ink is therefore attributed to the swollen and damaged TPU substrate rather than the ink itself. As illustrated in Figure 19(a), poor adhesion is observed on TPU due to its very low surface energy. Figure 19(b) further shows that this adhesion is further weakened after moisture exposure as a result of the hygroscopic behaviour of TPU. Together, these results indicate that varying terpinol content alone cannot compensate for TPU's low surface energy and thermal limitations, and that all TPU samples exhibit poor morphology, weak adhesion, and unreliable or unmeasurable electrical performance.

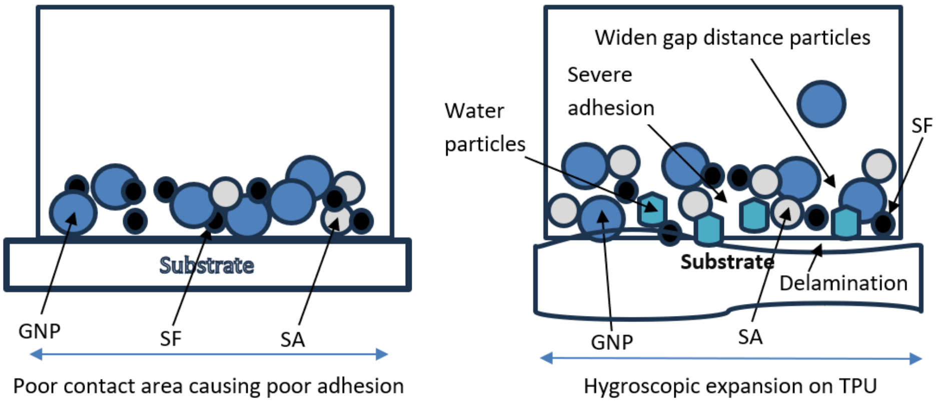


Figure 19. Adhesion for TPU (a) poor adhesion caused by low surface energy and (b) worsen adhesion caused by TPU hygroscopic properties after water immersion

4. CONCLUSION

The findings of this study highlight the substrate-dependent variability in the performance of GNP–Ag hybrid conductive inks. Among the investigated substrates, copper exhibited the best electrical performance, where the Cu 10T formulation achieved the lowest dry resistivity of approximately $\approx 1.2\times10^{-5}$ Ω.m, while Cu 15T showed the lowest wet resistivity of approximately $\approx 2.0\times10^{-5}$ Ω.m. These results are attributed to the formation of dense and well-adhered microstructures, making Cu 10T and Cu 15T the most suitable candidates for wet applications. In contrast, PET substrates exhibited higher resistivity values, reaching up to $\approx 10^{-4}$ Ω.m, along with clear performance degradation after immersion due to low surface energy, limited curing at 100 °C for one hour, and the formation of porous and discontinuous conductive networks. TPU substrates showed very high or unmeasurable resistivity after immersion, which is consistent with severe ink loss and hygroscopic swelling of the substrate. Although increasing terpinol content improved the initial film thickness and coverage, particularly for the 15T formulation, it also increased porosity and was insufficient to overcome poor wetting and moisture-induced deformation on PET and TPU. These observations clarify the dual role of terpinol in influencing both morphology and adhesion. Furthermore, the limitations of this study include the restricted low-temperature curing window for PET and TPU substrates, the use of a single SA particle size, and the consideration of only one immersion condition.

Overall, this work systematically investigated GNP/Ag/SA hybrid inks with 5T, 10T, and 15T terpinol on Cu, PET, and TPU substrates by comparing voltage, resistance, and resistivity under dry and wet conditions and by relating these results to surface energy, curing behavior, and hygroscopic effects. Future work should explore surface treatments for PET and TPU, broader optimization of solvent ratios, SA content and particle size, as well as long-term cyclic humidity, temperature, and mechanical testing to establish robust substrate–ink combinations for practical wet-environment applications.

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AUTHOR CONTRIBUTIONS STATEMENT

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C : **C**onceptualization
M : **M**ethodology
So : **So**ftware
Va : **Va**lidation
Fo : **Fo**rmal analysis
I : **I**nvestigation
R : **R**esources
D : **D**ata Curation
O : Writing - **O**riginal Draft
E : Writing - Review & **E**diting
Vi : **Vi**sualization
Su : **Su**pervision
P : **P**roject administration
Fu : **Fu**nding acquisition

CONFLICT OF INTEREST STATEMENT

No conflict of interest.

DATA AVAILABILITY

The data supporting the findings of this study are available within the article.

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