ELECTRICAL STABILITY UNDER MECHANICAL DURABILITY TESTS ON DIFFERENT SUBSTRATES

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Abstract:
With the ever-accelerating technological progress, there is an increasing awareness of more environmentally friendly materials and acceptance of the fact that the development of such materials in all fields of industry has become a necessity. The development of environmentally friendly conductive ink was the aim of this research, where the synthetic base was replaced with a natural one, given that the synthetic base, although water-based, still contains harmful compounds. Tapioca starch was proven to be a worthy replacement for synthetic base, and the conductive material graphite as a replacement for rare metals that are unsustainable, both environmentally and financially. Environmentally friendly conductive ink was prepared by homogenization on a magnetic stirrer with heating until the desired viscosity was achieved, i.e. gelatinization of tapioca starch, and after cooling, it was coated onto different substrates. Electrical conductivity of the printed samples was measured before and after mechanical tests to evaluate the durability and functional stability of the developed ink across different substrates. The development of the new ink formulation focuses on its application as a visible design element on packaging.

Keywords: *conductive ink, graphite powder, tapioca starch, electrical stability, mechanical durability*

1. INTRODUCTION

Environmentally friendly electronic systems imply the use of sustainable or biobased materials, or biodegradable and/or recyclable materials. Biodegradable electronics, commonly referred to as green electronics, remain a challenge since the composition of conductive inks must also be sustainable. A typical conductive ink consists of a functional material, a binder, a solvent, and optional additives combined to ensure electrical conductivity and stability [1], [2], [3]. Conductive inks are typically made from metallic (e.g., silver, copper), carbon-based (graphite, graphene, carbon nanotubes), or polymeric materials dispersed in a suitable binder and solvent. Graphite is inexpensive, chemically stable, and environmentally friendly. These characteristics make it a suitable material for printed and disposable electronics. It also offers sufficient electrical conductivity for flexible electronic applications[4], [5], [6]. Tapioca starch is a biodegradable and renewable biopolymer. It has good film-forming ability and flexibility, and it is low-cost. In conductive inks, it is used as a natural binder. It represents a sustainable alternative to synthetic polymers for eco-friendly printed electronic sensors. [7], [8], [9], [10].

In this study, a conductive ink based on graphite, tapioca starch, D-sorbitol, and water was investigated. The focus was on the electrical stability of the printed ink after mechanical durability tests on different substrates. Graphite provides electrical conductivity due to its layered carbon structure. Tapioca starch acts as a bio-based binder, forming cohesive films and helping to maintain particle dispersion during drying. D-sorbitol is added as a plasticizer to improve the mechanical and barrier properties of the starch-based film, particularly tensile strength and thermal stability. Water is used as the solvent, enabling safe and simple processing.

2. EXPERIMENTAL

Conductive ink containing 30% graphite powder was prepared by mixing 6 g of tapioca starch, 0.06 g of D-sorbitol (1% of the tapioca mass), and 24 g of fine graphite powder (Sigma-Aldrich, particle size < 20 µm). Distilled water was added to reach a total volume of 50 ml. The mixture was then heated to 80 °C on a magnetic stirrer with a heater (IKA RCT basic IKAMAG® safety control, Germany) until gelatinization occurred. After cooling to room temperature, additional graphite powder was gradually added while stirring continuously, resulting in a homogeneous ink.
Two types of paper were used as substrates: a coated paper (UPM Finesse White Gloss, 300 g/m²) and an uncoated bio-based paper (250 g/m²). The coated paper has a smooth, stable surface that helps prevent deformation during coating and allows for a more consistent evaluation of the ink's properties. The uncoated paper is made from a mix of 15% citrus-based agro-industrial by-products, 40% recycled fibres, and 45% virgin wood pulp, giving it a natural texture while keeping it environmentally friendly. The citrus fibres come from leftover fruit and juice-processing waste, supporting a circular use of resources.
The electrical stability of the printed coatings was tested using a simple circuit based on the Technic Conductivity Test Apparatus (Technic Inc., USA). A 4.5 V DC power source, an ammeter, a light-emitting diode (LED), and the printed coating were connected in series. Current flow was measured, and LED brightness served as a visual indicator of conductivity. Samples were tested before and after mechanical durability treatments, including rubbing and creasing. All measurements were performed at ambient laboratory conditions and repeated five times for reproducibility. The LED voltage drop was measured separately to determine the effective voltage applied to the print. Electrical resistance (R) was calculated using Ohm's law, and sheet resistance (R_s) was determined from the sample geometry.
The Hanatek Rub & Abrasion Tester was used to evaluate the rub resistance of the printed samples. Circular printed specimens (50 mm diameter) were placed in contact with paper substrates (115 mm diameter), and controlled rubbing was applied using two calibrated weights to achieve a total pressure of 2.0 psi. The tribometer operates with two rotating plates that maintain full contact, producing a uniform frictional motion. Each sample was subjected to 10 complete rotations, including areas of high ink coverage and fine printed lines, to observe differences in abrasion behaviour. The Zünd M-800 digital cutter was used to crease the printed samples at depths of 0.33, 0.66, 0.99, 1.33, 1.66, and 1.99 mm.
Microscopic analysis was performed using an Olympus BX51 optical microscope (Olympus, Japan) equipped with a DP72 digital camera (12.8 megapixels, 4140 × 3096 pixels). Images were acquired at 50× magnification for printed samples.

3. RESULTS AND DICUSSION

The following results summarize the electrical and mechanical behaviour of the conductive coatings printed on smooth coated and uncoated bio-based citrus substrates. The comparison between the two substrate types shows that the uncoated bio-based surface provided significantly better conductivity (lower sheet resistance) compared to the smooth coated paper (Table 1).

Table 1. Mean sheet resistance (R_s) of starch-based conductive coatings printed on different paper substrates

Substrate	Sample	Rs/$\Omega \cdot m^{-2}$
uncoated bio-based paper	U1	7.14 ± 0.03
	U2	3.32 ± 0.07
	U3	4.29 ± 0.02
	U4	6.99 ± 0.11
coated paper	C1	14.64 ± 0.06
	C2	12.82 ± 0.07
	C3	8.18 ± 0.03
	C4	10.81 ± 0.05

This difference can be attributed to the different wetting and absorption behaviour of the substrates. The uncoated citrus-based substrate likely promotes better particle packing and film continuity, resulting in denser conductive paths. In contrast, the coated paper's smoother, less absorbent surface may cause partial segregation of graphite particles during drying, leading to thinner or less uniform conductive layers.

Despite identical printing and formulation conditions, this result demonstrates that substrate surface chemistry and topography have a decisive influence on the final electrical properties of printed conductive coatings. The small SD values within each group confirm good repeatability, while the inter-group variation highlights the need to optimize substrate–ink interactions to achieve consistent performance.

The results of creasing at different depth (Table 2) show that the uncoated bio-based U4 sample showed a strong nonlinear response to mechanical creasing. At the smallest depth (0.33–0.66 mm), the sheet resistance increased drastically—up to nearly six times the initial value—indicating disruption of the conductive network caused by microcracking and surface detachment of the printed layer.

Table 2. Average sheet resistance (R_s) of starch-based graphite conductive coatings after creasing at different depth (0.33, 0.66, 0.99)

Creasing depth (mm)	Rs(U4)/ $\Omega \cdot m^{-2}$	U4 (Change) /%	Rs(C3)/ $\Omega \cdot m^{-2}$	C3 (Change) /%
0.00	4.29 ± 0.02	—	8.18 ± 0.03	—
0.33	24.47 ± 1.12	470.0 %	26.38 ± 0.39	222 %
0.66	28.85 ± 0.80	572 %	26.29 ± 0.18	221 %
0.99	18.70 ± 0.58	336 %	31.04 ± 0.17	280 %

However, at a depth of 0.99 mm, resistance partially decreased, which suggests partial recovery of conductivity due to elastic relaxation and reconnection of conductive particles within the composite matrix. In contrast, the smooth coated substrate (C3) exhibited a monotonic increase in sheet resistance throughout creasing, reaching almost three times the initial value at the largest depth. This steady degradation points to progressive crack propagation and loss of interparticle contact, characteristic of a stiffer, less compliant surface that does not permit elastic recovery.

Table 3. Average sheet resistance (R_s) of starch-based graphite conductive coatings after creasing at different depth (1.33, 1.66, 1.99)

Creasing depth (mm)	Rs(U2)/ $\Omega \cdot m^{-2}$	U2 (Change) /%	Rs(C2)/ $\Omega \cdot m^{-2}$	C2 (Change) /%
0.00	3.32 ± 0.06	—	12.82 ± 0.07	—
1.33	6.13 ± 0.01	+84.6 %	10.51 ± 0.04	−18.0 %
1.66	4.58 ± 0.02	+37.9 %	12.22 ± 0.18	−4.7 %
1.99	4.24 ± 0.02	+27.7 %	14.90 ± 0.20	+16.2 %

For the smooth substrate (C2) (Table 3), the sheet resistance initially decreased upon creasing to a small depth (1.33 mm), likely due to temporary compression of the conductive graphite network and improved interparticle contact. However, further creasing led to an increase in resistance, exceeding the initial value at 1.99 mm, which indicates mechanical degradation of the conductive layer through the formation of microcracks and partial loss of continuity. This trend reflects an elastic–plastic response, where initial deformation enhances conductivity before structural damage dominates.

In contrast, the uncoated citrus-based substrate (U2) exhibited an opposite trend. A sharp initial increase in sheet resistance (≈ +85%) was observed at the smallest creasing depth, suggesting crack initiation or delamination between the conductive layer and the less uniform bio-based surface. As the creasing depth increased, the resistance gradually decreased, implying partial re-establishment of conductive pathways due to relaxation of surface strain and elastic recovery of the substrate.

The results indicate that the substrate type strongly influences the mechanical–electrical stability of the printed conductive film. The smooth coated paper provided a more stable and uniform conductive response, while the uncoated citrus-based substrate demonstrated greater flexibility and recovery potential, though at the cost of higher initial sensitivity to deformation.

The results showed that the uncoated citrus-based substrates had higher mechanical flexibility. They also displayed partial recovery of conductivity after creasing, reflecting their elastic and compliant surface characteristics. The smooth coated papers, in comparison, showed greater structural stability but a progressive loss of conductivity. This indicates that substrate stiffness limits the mechanical–electrical resilience of printed conductive layers.

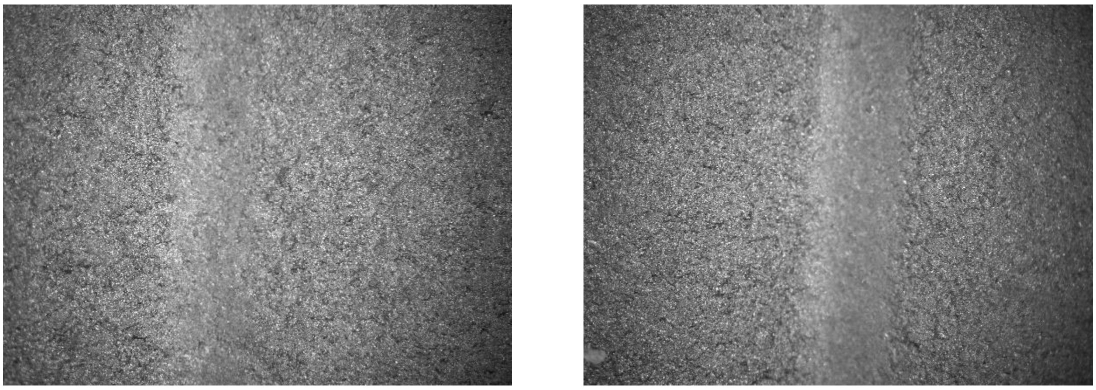
Table 4. Sheet resistance of printed conductive layers before and after abrasion test

Substrate	Condition	Rs/ $\Omega \cdot m^{-2}$	Change/%
Uncoated bio-based paper	Before abrasion	6.31 ± 0.45	—
	After abrasion	8.18 ± 0.02	+29.7 %
coated paper	Before abrasion	14.64 ± 0.09	—
	After abrasion	14.59 ± 0.04	−0.34 %

The mechanical–electrical characterization revealed distinct behaviors of the conductive coatings depending on substrate type (Table 4).

During creasing tests, uncoated citrus-based substrates showed higher flexibility and partial recovery of conductivity. This suggests an elastic and compliant interface that allows reconnection of conductive particles after deformation. However, under rubbing, these same samples showed an increase in sheet resistance (≈30%). This indicates that their surface cohesion is lower, and the conductive network is more sensitive to abrasion and surface disruption.

Conversely, smooth coated paper substrates displayed greater dimensional and structural stability. Their sheet resistance remained nearly unchanged after rubbing (−0.3%), suggesting good mechanical integrity and strong adhesion of the conductive layer. Nevertheless, creasing caused a progressive increase in resistance, implying microcrack formation within the stiffer, less elastic surface.



a) Uncoated bio-based paper b) coated paper
Figure 1. Surface morphology of creased conductive coatings printed on uncoated bio-based and coated paper substrates at 50× magnification

Microscopic examination (Figure 1) showed differences in the surface of the conductive coatings after creasing. On the uncoated citrus-based paper, small microcracks and partial layer deformation were observed. Most of the conductive film stayed intact, indicating good flexibility and partial recovery of the conductive network upon creasing. The coated paper exhibited more noticeable cracking and flake detachment, reflecting its stiffer surface and limited ability to deform elastically. These observations suggest that the electrical and mechanical results are consistent with the idea that substrate texture and flexibility play a role in crack formation, adhesion, and the stability of the conductive layers under mechanical stress.

Visual inspection after the abrasion test showed difference between the two substrates. The uncoated citrus-based paper showed a transfer of graphite onto the white sheet, indicating weaker surface cohesion and higher material loss during rubbing. The coated paper showed only slight grey traces with minimal visible damage on the rubbed area, suggesting better adhesion and abrasion resistance of the conductive layer.

4. CONCLUSION

The results suggest that substrate morphology and mechanical properties influence the performance of starch–graphite conductive coatings. The uncoated bio-based citrus paper tended to have lower sheet resistance and offered greater flexibility. It also showed reduced surface cohesion and was more sensitive to abrasion. The smooth coated paper provided better adhesion and mechanical stability, although conductivity gradually decreased when the surface was creased. Microscopic observations of creased surfaces supported these trends, showing crack patterns that matched the observed electrical behavior. The balance between conductivity, flexibility, and durability seems to depend largely on the properties of the substrate surface. Future research should explore changes to the ink composition or surface modifications to improve adhesion and resistance to mechanical stress, while still maintaining good electrical performance and environmental friendliness.

5. ACKNOWLEDGEMENT

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