

Eco-Friendly, High-Performance Cellulose Acetate Encapsulation for Flexible Hybrid Electronics: Environmental Stability and Printing Scalability

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Abstract

In this study, we developed a screen-printable, cost-effective, biocompatible, and biodegradable dielectric ink based on cellulose acetate for encapsulating biomedical electronic device packaging. To evaluate its environmental stability, the encapsulated samples were subjected to accelerated aging conditions of 70 °C and 80% relative humidity for 216 hours. Three groups were tested: non-encapsulated controls, cellulose acetate-encapsulated samples, and samples encapsulated with a commercial material (Micromax ME778). Sheet resistivity was measured before and after environmental aging to assess the protective performance of each encapsulant. The cellulose acetate-encapsulated samples exhibited only an 11.8% increase in sheet resistivity, compared with a significant 65.6% increase observed in the ME778 group. These results demonstrate the superior environmental protection and suitability of cellulose acetate as a sustainable encapsulant for printed biomedical electronics.

Keywords

Biodegradable, cellulose acetate, encapsulant, printed electronics, screen-printing, temperature and humidity.

I. Introduction

Additive manufacturing has revolutionized electronics packaging by enabling the production of complex, customized components with high precision [1]. It supports a wide range of materials, including conductive inks for circuit patterns and encapsulants for device protection [2].

Effective encapsulation is critical in electronics packaging, as it protects devices from external stresses such as humidity, dust, corrosion, electrical interference, and mechanical strain [3]. Consequently, it directly impacts device performance, reliability, and operational lifetime [4]. Material selection plays a key role in achieving this protection because it determines the encapsulant's mechanical strength, chemical stability, and environmental resistance [5],[6]. Since encapsulation directly influences performance and longevity, the chosen materials must not only provide environmental protection but also be compatible with roll-to-roll (R2R) fabrication and sustainability requirements [7]–[10].

With the growth of flexible electronics in biomedical applications, such as wearable electrocardiogram (ECG)

devices, there is an increasing demand for materials compatible with additive and R2R manufacturing processes to enable high-throughput, cost-effective production of biocompatible devices [11]–[13]. At the same time, the rise in hazardous electronic waste has created a strong need for eco-friendly and biodegradable materials, particularly for single-use applications [14]. Furthermore, the use of low-cost materials supports mass production and contributes to the development of affordable final products.

Flexible electronic devices, particularly in wearable and disposable formats, are increasingly used for continuous health monitoring and home-based care [15]. These devices require encapsulants that can reliably withstand high humidity and elevated temperatures to ensure consistent signal quality and maintain material integrity over extended shelf life.

Common challenges associated with encapsulation materials include performance degradation under elevated temperature and humidity conditions, lack of sustainability, and limited scalability [4]. To address these issues, we investigated cellulose acetate, a naturally derived form of cellulose known for its biocompatibility, biodegradability, chemical

tunability, and cost-effectiveness, as a potential encapsulation material for biomedical electronics [16]. This material offers a promising solution to many of the challenges outlined above.

Cellulose acetate is a chemically modified form of cellulose in which some hydroxyl groups are replaced with acetyl groups. This modification reduces water absorption and enhances thermal stability, making it suitable for encapsulation applications. Beyond encapsulation, cellulose acetate is also used in drug delivery systems as a drug carrier, in gas and ion separation membranes, and in food packaging [17]–[19].

Furthermore, formulating cellulose acetate into a screen-printable ink enhances its scalability for industrial applications. Among various additive manufacturing techniques, screen printing is particularly advantageous for fabricating flexible electronics. In screen printing, a stencil (or screen) is used to transfer ink onto a substrate, creating patterns or layers with high accuracy. This method is widely employed in electronics for its simplicity, cost-effectiveness, and scalability. Its compatibility with flexible substrates makes it ideal for large-scale production of flexible, wearable, and biomedical devices. Additionally, screen printing supports high-throughput manufacturing, which is essential for mass production in the growing field of flexible hybrid electronics [20].

In this study, we developed a screen-printable, cellulose acetate-based encapsulant and evaluated its performance under elevated humidity and temperature conditions, comparing it with a commercial encapsulant and non-encapsulated control samples.

Our findings demonstrate that the cellulose acetate-based encapsulant provides superior environmental stability, exhibiting only an 11.8% increase in sheet resistivity after 216 hours of exposure to 70 °C and 80% relative humidity, compared with a 65.6% increase observed in samples encapsulated with the commercial Micromax ME778 material. These results highlight the potential of cellulose acetate encapsulant for scalable and sustainable use in biomedical electronics.

II. Materials and Methods

The experimental process began with the preparation of the cellulose acetate-based ink, followed by viscosity measurements to ensure suitable rheological properties for screen printing. The prepared ink was then printed onto substrates, and resistance measurements were performed both before and after encapsulation to assess the impact of the protective layer on electrical properties. To further evaluate the stability and robustness of the printed structures, samples were subjected to environmental aging tests under controlled temperature and humidity conditions. Subsequent

sheet resistivity analysis was conducted to quantify any changes in electrical performance over time. This sequence of experiments provided a comprehensive evaluation of the ink's compatibility with the printing process and its long-term stability under operational conditions (Fig 1).

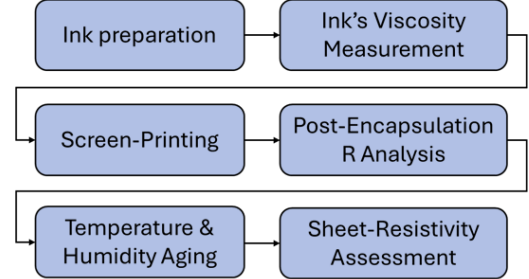


Figure 1: Overview of the experimental procedure

A. Ink Preparation

To prepare the cellulose acetate-based encapsulation ink, cellulose acetate (Product No. 419028, average Mn ~50,000 by GPC, Sigma-Aldrich, St. Louis, MO, USA) was mixed with ethylene glycol diacetate (99%, Product No. 525200, Sigma-Aldrich, St. Louis, MO, USA), deionized (DI) water, and ethanol in optimized ratios at room temperature (Table I).

Table I: Cellulose acetate ink formulation

	Cellulose acetate	Ethylene Glycol Diacetate	DI water	Ethanol
Weight (g)	1	5	1	1

B. Viscosity Measurement

The viscosity of the prepared cellulose acetate-based encapsulation ink was measured using a DVNext cone/plate rheometer (Brookfield, Middleboro, MA, USA). For each measurement, 500 μ L of the ink was tested at room temperature using rotational speeds of 0.4, 0.3, and 0.25 rpm. The measured viscosity values ranged from 21,170 to 25,690 cP, which is well-suited for screen-printing applications (Fig 2A).

C. Dielectric Constant Measurement

The dielectric constant of the prepared cellulose acetate-based encapsulation ink was measured using a dielectric assessment kit for thin layers (DAK-TL2) with the open coaxial probe method. The instrument was calibrated, and the accuracy of the calibration was verified prior to the dielectric constant measurements. A printed sheet of the encapsulation ink was fabricated for this purpose. Four independent measurements were performed, yielding highly consistent results with negligible standard deviation (Fig 2B). The measured dielectric constant fell within the expected range for cellulose acetate.

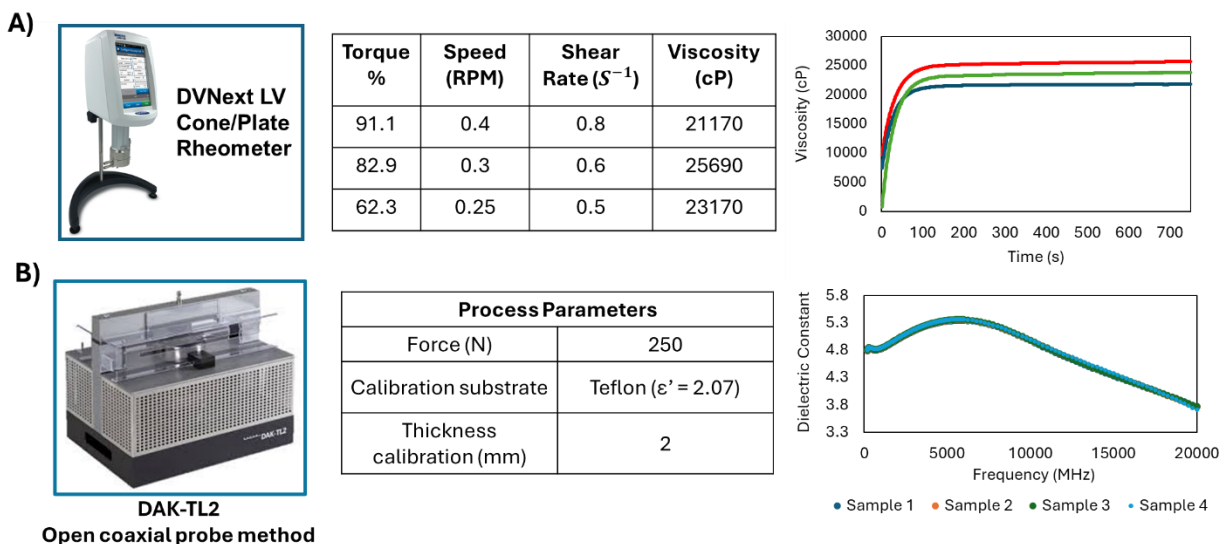


Figure 2: (A) Measured dielectric constant values for four cellulose-based encapsulation materials. (B) Viscosity range of the prepared cellulose acetate-based ink measured at different shear rates. Three independent measurements were conducted at room temperature to confirm suitability for screen-printing applications.

D. Screen-printing

After preparation, the cellulose acetate-based encapsulation ink was screen-printed onto previously fabricated carbon electrode leads on 5 mil Kapton polyimide substrates using an MPM screen printer. The printing parameters were optimized specifically for the cellulose acetate ink formulation to achieve uniform, defect-free coverage. This process resulted in an average encapsulation layer thickness of approximately $4.5\ \mu\text{m}$, as confirmed by profilometry measurements.

The electrode leads were fabricated in varying widths to investigate the impact of geometry on electrical performance (Fig 3A). As shown in Fig 3D, wider electrode linewidths resulted in lower resistance, consistent with the expected increase in conductance due to the larger cross-sectional area. To systematically evaluate the effect of encapsulation, the printed electrode sheets were divided into three groups, each containing a minimum of four independent samples: (i) electrodes without encapsulation, (ii) the electrodes encapsulated with a commercially available encapsulant (Micromax ME778), which contains titanium dioxide (40–50 wt%) and dipropylene glycol methyl ether (30–40 wt%), and (iii) electrodes encapsulated with the cellulose acetate-based material (Fig 3A). This grouping enabled a direct comparison of the effects of encapsulation materials on the electrical and environmental stability of the electrodes.

After printing, the cellulose acetate encapsulant was cured in a reflow oven at the optimized temperature of $120\ ^\circ\text{C}$ for 6 minutes. This curing step was chosen to ensure complete solvent evaporation and the formation of a uniform, adherent film without compromising the underlying electrode structure or substrate integrity. Following curing, the printed

and encapsulated electrodes were allowed to cool to room temperature before subsequent electrical and environmental testing.

III. Results and Discussion

A. Effect of Encapsulation on Electrodes' Resistance

The electrical resistance of the electrode leads was measured before and after encapsulation using a Keithley 2100 digital multimeter to evaluate the effect of encapsulation on the electrodes' electrical performance. Encapsulation with the cellulose acetate-based ink resulted in a resistance increase of 5.2–11.7%, corresponding to electrode linewidths from 10 mm to 1 mm, respectively, indicating minimal disruption of the conductive pathways. In contrast, electrodes encapsulated with the commercially available Micromax ME778 material exhibited a substantially higher resistance increase of 49.8–63.1% (Fig 3D). The observed differences can be attributed to the distinct interactions between the encapsulant materials and the printed graphene-based electrodes, including factors such as solvent compatibility, penetration into the electrode structure, and adhesion characteristics. These results suggest that the cellulose acetate encapsulant is more suitable for graphene-based inks, as it preserves the electrical conductivity of the electrodes while providing the desired protective coverage. Overall, the data demonstrate that careful selection of encapsulation material is critical for maintaining device performance in flexible printed electronics applications.

Scanning electron microscope (SEM) imaging was performed on the surface of screen-printed carbon electrodes, as well as samples encapsulated with cellulose

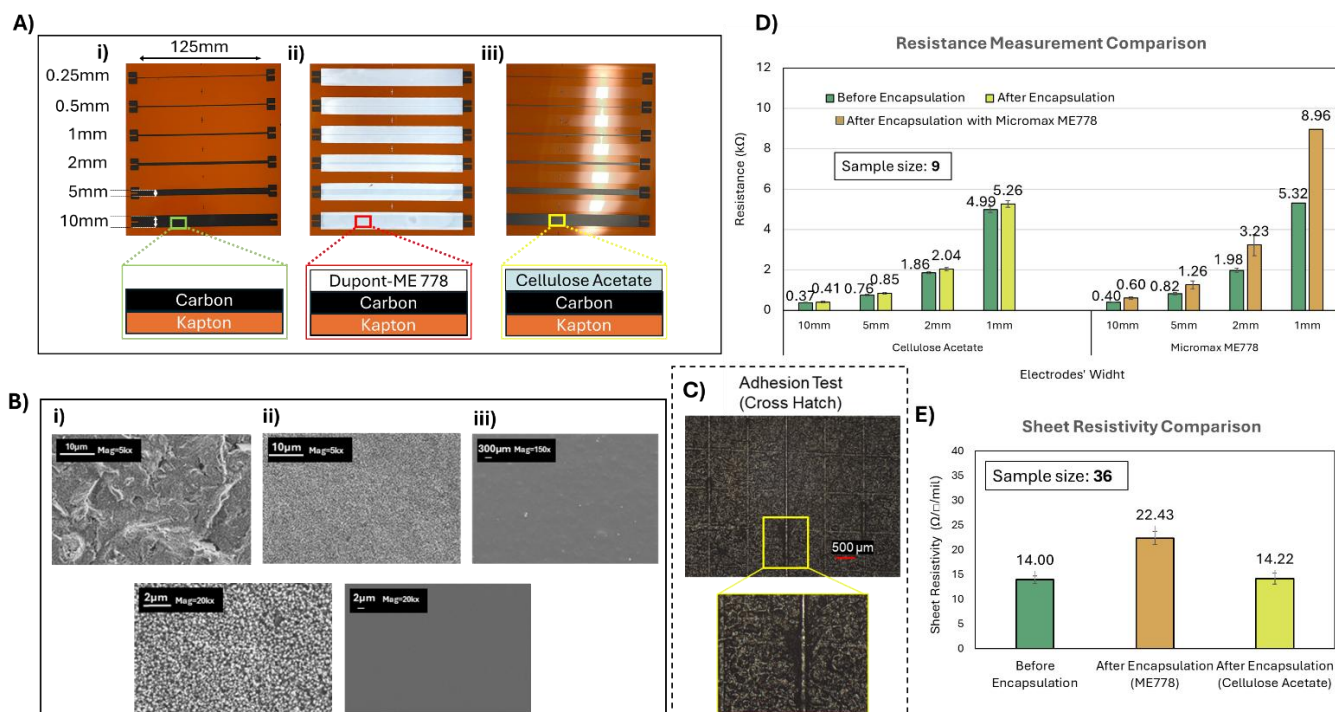


Figure 3: (A) Screen-printed electrode leads on Kapton substrate: (A-i) Non-encapsulated controls with indicated dimensions; (A-ii) Electrode leads encapsulated with the commercial encapsulant Micromax ME778; (A-iii) Electrode leads encapsulated with the developed cellulose acetate-based encapsulant. (B) SEM images of: (B-i) screen-printed carbon electrode lead; (B-ii) Micromax ME778-encapsulated carbon electrode at magnifications of 5kx and 20kx; (B-iii) cellulose acetate-encapsulated carbon electrode at magnifications of 150x and 20kx. (C) Cross-hatch test result of the cellulose acetate encapsulant showing excellent adhesion with only minimal detachment. (D) Comparison of electrode resistance before and after encapsulation using Micromax ME778 and cellulose acetate-based inks. (E) Sheet resistivity comparison of samples before encapsulation and after encapsulation with ME778 and cellulose acetate-based ink.

Scanning electron microscope (SEM) imaging was performed on the surface of screen-printed carbon electrodes, as well as on samples encapsulated with cellulose acetate and Micromax ME778 inks. SEM images were acquired using a Gemini Supra SEM and a Hitachi SEM for different sample sets (Fig 3B).

SEM images taken at various magnifications provided detailed insights into the surface morphology of the encapsulation materials. The Micromax ME778 encapsulant exhibited a highly porous structure, whereas the cellulose acetate-based encapsulant showed a uniform, dense polymer layer with no visible porosity. This uniform coverage suggests that the cellulose acetate encapsulant forms a more effective barrier against environmental factors such as humidity, supporting its superior performance observed during high-temperature and high-humidity aging tests.

B. Cellulose Acetate Encapsulant Adhesion Assessment

The adhesion of the cellulose acetate-based encapsulant was evaluated using an Elcometer 1542 Cross Hatch Adhesion Tester. A cross-hatch pattern was created by pressing and

pulling the cutter wheel in two perpendicular directions.

Adhesion tape was then applied to the patterned area and carefully removed. The tested surface was inspected using a Keyence microscope. Imaging revealed less than 5% impact from the test, with only minimal peeling observed at the corners of the squares (Fig. 3C). These results confirm the excellent adhesion of the cellulose acetate encapsulant on the carbon-based ink.

C. Temperature and Humidity Testing

Screen-printed samples, including those encapsulated with cellulose acetate, ME778, and non-encapsulated controls, were subjected to accelerated aging under high humidity and temperature conditions. The samples were exposed to 70 °C and 80% relative humidity for 216 hours in a Tenney Environmental Test Chamber. This accelerated aging protocol simulates long-term operational stresses and allows assessment of the encapsulants' protective performance over time. To ensure the reliability and reproducibility of the results, a minimum of three samples per group were tested (Fig 4).

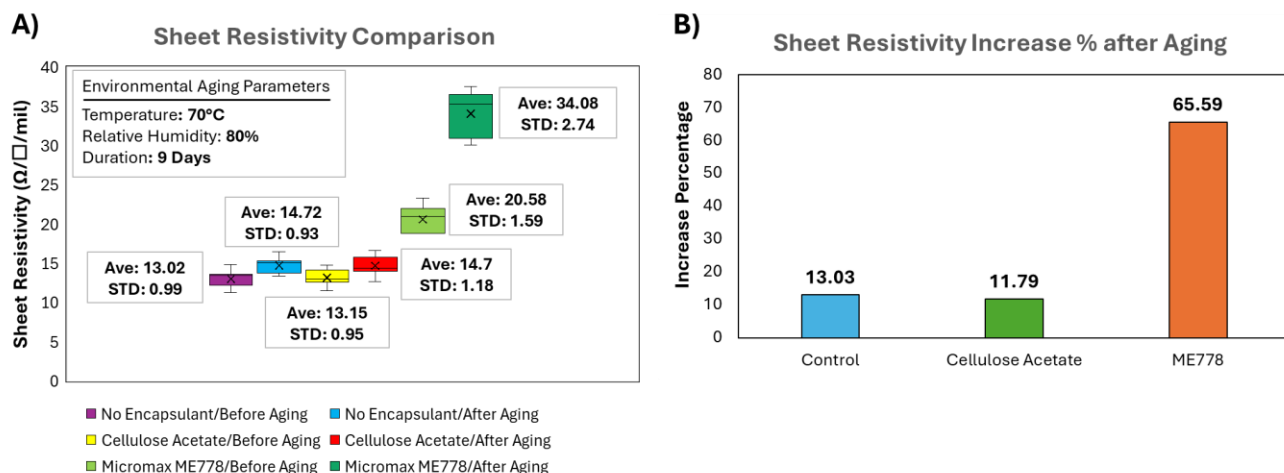


Figure 4: (A) Comparison of sheet resistivity for encapsulated samples (Micromax ME778 and cellulose acetate) and non-encapsulated controls before and after temperature and humidity aging. (B) Percentage change in sheet resistivity following environmental aging at 70 °C and 80% relative humidity for 216 hours.

The results demonstrated that the developed cellulose acetate-based encapsulant effectively protected electrode leads from elevated humidity and temperature, outperforming both the non-encapsulated control samples and those encapsulated with the commercial material (Micromax ME778). After 216 hours of exposure to 70 °C and 80% relative humidity, the sheet resistivity of the cellulose acetate-encapsulated samples increased by only 11%, indicating stable and reliable performance. In comparison, the control samples showed a 13% increase, while the ME778-encapsulated samples exhibited a substantial 65% increase in sheet resistivity. The inferior performance of the ME778 encapsulant compared to the cellulose acetate-based material suggests that the commercial encapsulant was less effective under accelerated environmental aging conditions, potentially due to material degradation or compromised encapsulant integrity. The highly porous morphology of ME778, as observed in SEM imaging, may facilitate rapid moisture ingress, leading to swelling, loss of adhesion, and accelerated degradation of the conductive pathways. In contrast, the dense and uniform polymer layer formed by cellulose acetate acts as an effective barrier against both moisture penetration and thermal stress, thereby maintaining the structural integrity of the encapsulant and ensuring stable electrical properties during aging.

IV. Conclusion

The developed cellulose acetate-based dielectric ink was successfully screen-printed, demonstrating excellent compatibility with graphene-based printed electrodes. Electrical measurements revealed only a modest increase in resistance (5.2–11.7%) after encapsulation with the cellulose

acetate ink, compared to a significantly higher increase (49.8–63.1%) observed with the commercial Micromax ME778 encapsulant. Environmental aging tests conducted at 70 °C and 80% relative humidity for 216 hours confirmed the suitability of the cellulose acetate encapsulant for withstanding challenging environmental conditions, showing only an 11% increase in sheet resistivity compared to a 65% increase observed in samples encapsulated with ME778. These results highlight the material's potential to significantly enhance the longevity and reliability of flexible devices, making them an excellent candidate for biomedical applications. In addition to its electrical and environmental advantages, the material's screen-printability and compatibility with R2R manufacturing make it a cost-effective and scalable solution for industrial applications. The encapsulant's biocompatibility, biodegradability, and environmental stability further underscore its promise for advancing sustainable biomedical device technologies while addressing the growing need for reliable, eco-friendly materials.

Future Work

While the current study demonstrates the environmental stability of the cellulose acetate-based encapsulant under elevated temperature and humidity conditions, further investigation is needed to assess its mechanical reliability. Future work will focus on evaluating the encapsulant's performance under mechanical stresses, such as bending, which are commonly encountered in wearable biomedical devices. In addition, thermocycling tests will be performed to better simulate real-world operating conditions. These experiments aim to provide deeper insight into the long-term

adhesion, structural integrity, and electrical stability of the encapsulant, thereby supporting its potential for use in practical flexible and wearable electronic applications.

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