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REGULAR PAPER

Highly Flexible, Conductive, and Antibacterial Surfaces Toward Multifunctional Flexible Electronics

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Abstract

Conductive metallization of polymer surfaces, owing to the integration of unique features of dissimilar materials (i.e., polymer + metal), is becoming the central focus in flexible polymer electronics. However, fabrication of multifunctional surfaces on polymers in a high-throughput and robust manner at ambient conditions remains challenging. In this study, we employ the cold spray (CS) particle deposition technique to produce multifunctional hybrid surfaces on a flexible polymeric substrate (PET) toward flexible electronics. In this regard, soft metal particles (Sn), are deposited on the polymer surface as an "interlayer" followed by the over-coating of hard metal (Cu) film to create hybrid (Sn + Cu) surfaces. Studies on microstructure, adhesion strength, and water contact angle are conducted to characterize the resulting surface structure. By leveraging the optimum CS settings, multifunctional surfaces with promising electrical conductivity (5.96×10^5 S.m^{-1}), flexibility, adhesive strength, and hydrophobicity (contact angle $\approx 122°$) were achieved. Moreover, the antibacterial performance of the surface is confirmed by the in vitro antibacterial tests in a manner that > 99% of the bacteria were inhibited. This work provides a promising strategy for high-throughput manufacturing of multifunctional surfaces (flexible + conductive + antibacterial surfaces) toward multifunctional flexible electronics.

Keywords Multifunctional surface · Cold spray · Flexible electronics · Polymer metallization · Antibacterial coating

1 Introduction

Polymers have been widely used in numerous applications owing to their advantageous features, including, lightweight, high specific strength, formability, and corrosion resistance [1, 2]. In particular, flexible polymers, such as—poly(ethylene terephthalate) (PET), polyethylene naphthalate (PEN), polyimide (PI), polydimethylsiloxane (PDMS)—have become vital for next-generation soft and flexible printed electronics [3]. However, some major limitations of these polymers (e.g., dielectric nature, poor erosion resistance, low thermal budget, vulnerability to UV rays damage) impede their practical use in many electronics applications [1–4]. To address these challenges, functional surface metallization on these important materials seems to be an effective solution, allowing improved physical and mechanical surface properties [5].

Conventional metallization techniques of polymers mainly involve vapor deposition [1, 6], electroplating [4, 7], electroless plating [1, 8], screen printing [9], sputter coating [10, 11], aerosol jet printing (AJP) [12, 13], laser-induced

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metallization [14], and thermal spray [7, 15]. Besides their advantages, these methods face challenges such as low deposition rate (e.g., vapor deposition), high processing and equipment cost (e.g., laser-induced metallization), high process temperature (e.g., thermal spray), and sample size (in case of sputter coating, electroplating, electroless plating) [1, 6, 7, 15]. In particular, high-process temperatures often lead to the degradation of polymer structure, resulting in undesired porosity, oxidation, and surface distortion [6, 15, 16].

Recently, the cold spray (CS) particle deposition technique has emerged as a promising polymer metallization technique at low process temperatures [16–19]. The CS technique, owing to the solid-state deposition of particles, offers many opportunities for high-throughput functional metallization with minimal risk of oxidation and degradation [20]. In the CS, fine metal powders (5–50 µm) are accelerated to high velocities (300–1200 m/s) through a converging–diverging nozzle using compressed gases. Once the particles collide on a target surface, the particles are metallurgically bonded onto the substrate surface, creating a solid-state functional coating [20, 21]. Owing to its low heat input, CS has the potential for metallization of temperature-sensitive materials such as polymers and polymer composites [1, 22]. Particularly, most of the polymers can be metalized by CS using functional feedstock materials [e.g., copper (Cu), zinc (Zn), aluminum (Al), tin (Sn), iron (Fe), and silver (Ag)] to achieve improved surface features in terms of electrical, mechanical, and thermal properties, as well as antibacterial activity [1, 15]. As such, the CS offers promising potential to fabricate multifunctional surfaces for the state-of-the-art field of printed flexible electronics. In the current situation, however, it remains challenging to fabricate multifunctional surfaces on flexible polymers by the CS due to: (i) difficulty in deposition of harder metal particles (e.g., Cu) on intrinsically soft polymers [23]; and (ii) erosion of polymer substrate during the high-speed impact of particles [1, 23, 24]. Addressing these limitations is crucial for the pragmatic use of this emerging surface deposition technology in creating multifunctional flexible electronics.

In the present study, to develop multifunctional surfaces for flexible electronics, we address the above-mentioned challenges of the CS technique through the "inter-layer" strategy. In this regard, first, soft metal powders (i.e., Sn) are deposited on the flexible polymer target (PET) to achieve an electrically conductive initial surface. Next, the pre-deposited coating is utilized as an inter-layer for the subsequent Cu over-coating to gain antibacterial functionality to the pre-deposited Sn surface. In this way, the resulting hybrid metal (Sn + Cu) surface attributes multifunctionality with improved conductivity and antibacterial activity. The fabricated surfaces are thoroughly characterized in terms of microstructure, film thickness, electrical conductivity, adhesion strength, and surface wettability. The flexibility, electrical performance, and stability of the resulting surface are also tested under mechanical deformations such as bending and twisting. Moreover, in vitro antibacterial tests are conducted to characterize the antibacterial activity of the fabricated surface.

2 Material and Methods

2.1 Materials

A thermoplastic polymer, PET sheet (McMaster-Carr, USA, thickness = 0.25 mm), was used as a flexible substrate material due to its inherent advantages including low-cost, low surface-roughness, and recyclability [3]. Micron-scale Sn (10–45 µm) [25] and Cu (5–45 µm) [8, 26] feedstock particles were procured (Centerline, U.S.) and used as received. The morphology of Cu and Sn powders are shown in Fig. 1a, b, respectively. Sn was selected as the inter-layer coating material owing to its soft nature, corrosion resistance, and cold sprayability on various polymers [27–29]. Cu was used for over-coating to improve the surface properties in terms of hydrophobicity and antibacterial functionality owing to its superior antibacterial characteristics [30–32].

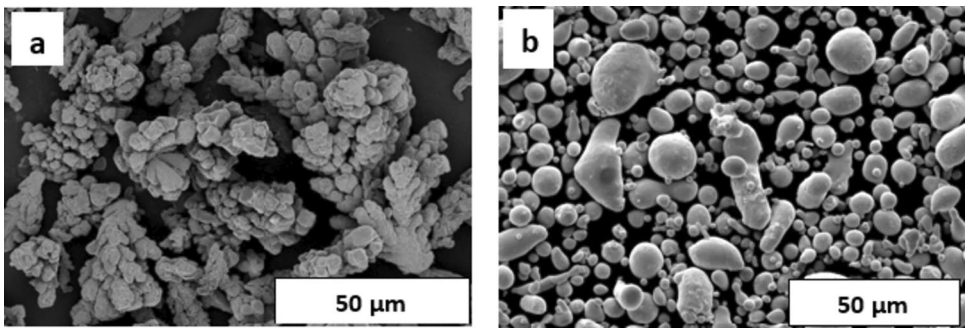


Fig. 1 SEM images of the feedstock powders: **a** Cu and **b** Sn powders

2.2 Cold Spray Surface Deposition

The CS is an emerging green surface deposition (metallization) technology that allows the creation of functional coatings on various substrates with improved bond strength at low-process temperatures [25, 33], offering a sustainable and environmentally friendly alternative to traditional manufacturing processes [34]. In this technique, particles are accelerated to supersonic velocities (300–1200 m/s) through a compressed gas flow by using a converging–diverging nozzle. When the particles impact the target surface, particles kinetic energy disperses on the surface, resulting in a high-strength functional coating [25, 35].

Unlike conventional methods that often involve high temperatures and produce hazardous emissions, CS utilizes kinetic energy to deposit particles onto a substrate at supersonic speeds without the need for melting. The solid-state nature of the CS technique minimizes energy consumption, reduces waste generation, and eliminates harmful by-products, making it an eco-friendly option for functional surface manufacturing [36]. Additionally, CS enables the use of recyclable materials, such as powders derived from waste products or reclaimed metals, further promoting sustainability. By facilitating efficient and cost-effective material usage and minimizing environmental impact, CS technology plays a vital role in advancing green manufacturing practices and supporting efforts towards large-scale functional surface coating in an environmentally friendly and sustainable manner [3, 34, 37].

Owing to the aforementioned advantages, the CS technique has recently gained great attention in functional metallization on polymeric substrates. Despite great promises, metallization on intrinsically soft polymers using hard feedstock particles (e.g., Cu) through the CS at low-process temperatures remains challenging [23]. The main reason is the severe porosities due to substrate erosion under the high-speed impingement of the particles [7]. As such, this phenomenon impedes stable electrical conductivity due to the discontinuous Cu deposition on polymer surfaces via CS [7, 8].

One potential solution to this challenge can be the utilization of the "inter-layer" strategy as illustrated in Fig. 2. In this approach, an initial layer of soft metal, such as Sn, is deposited on the polymer surface to fabricate an electrically conductive layer. Next, the as-deposited Sn is used as the "inter-layer" for the subsequent over-coating with hard particles such as Cu. In this way, hybrid (Sn + Cu) surfaces can be fabricated, comprising both electrical conductivity from the initial Sn deposit as well as antibacterial functionality from the Cu over-coating.

In this regard, Sn and Cu particles were sequentially deposited on the flexible PET polymer substrate to achieve a multifunctional hybrid surface (see Fig. 2). A low-pressure cold spray machine (Rus Sonic K205/407R) was used for the surface deposition experiments. The details of the CS experimental setup can be found in Akin et.al. [25]. Here, Sn powders are sprayed using an axisymmetric nozzle (outlet diameter = 4.5 mm) while Cu over-coating was performed with a rectangular-shaped nozzle with a larger exit aperture (i.e., 3 mm × 10.35 mm). This configuration allows the processing of a wider area of Cu-over coating on the pre-deposited Sn layer without any alignment issues. That said, in this arrangement, Sn coating provides electrical conductivity as the Cu over-layer improves the hydrophobicity and

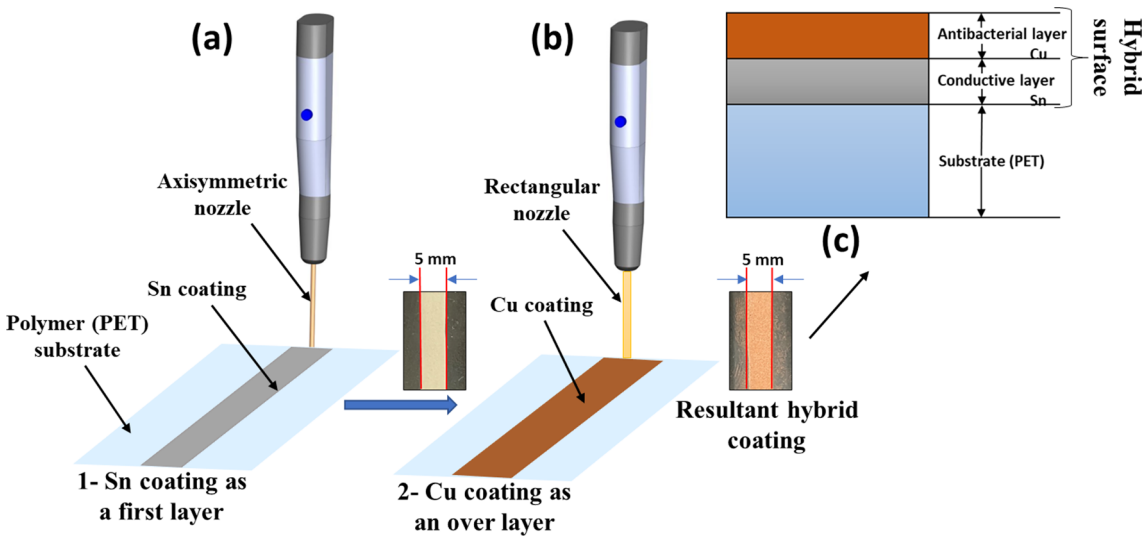


Fig. 2 Schematic of the manufacturing steps of the multifunctional surface via the CS of: **a** Sn inter-layer, **b** Cu over-coating; **c** cross-sectional schematic view of the hybrid surface

antibacterial properties, enabling a multi-functional flexible surface. The CS was performed using the experimental settings listed in Table 1, in which the settings of Sn coating were adopted from our previous work [25].

2.3 Characterization Methods

The electrical resistance of the surface was measured by the two-point probe method using a digital multimeter (Agilent/HP 34401A) while the sheet resistance properties as analyzed by a four-point probe system (Jandel, RM3-AR). A digital micrometer (REXBETI) with a resolution of 1 µm was used to measure the coating film thickness. The surface morphology, cross-section, and elemental analysis of the resultant coatings were conducted by a scanning electron microscope (SEM) equipped with an X-ray (EDX) detector (Hitachi S-4800 Field Emission SEM). To evaluate the adhesion strength of the hybrid coating, cross-cut adhesion tests were conducted based on the ASTM D3359 standard [38]. The surface wettability was studied by calculating the water contact angle based on the sessile drop method [39]. Lastly, the antibacterial characteristics of the resultant hybrid coatings were evaluated according to the ISO 221961 standard ('Plastics—Measurement of antibacterial activity on plastic surfaces') [40]. Three specimens (n = 3) were used in each characterization study at room temperature.

3 Results and Discussion

3.1 Characterization of the Cold Spray Surface Deposition

To investigate the effect of the CS settings (i.e., nozzle speed, spray distance, number of spray passes), a full factorial experimental design was constructed (See Appendix A1). Three variables at three levels were used to design the experimental studies. The factors are: (i) nozzle speed (50, 100, 200 m/s); (ii) spray distance (10, 30, 50 mm); and (iii) number of the spray pass (1, 3, 5 passes), while the outputs are electrical resistance (Ω) and coating film thickness (µm).

Table 1 Operational settings of the CS

Parameters	Sn coating	Cu over-coating
Driving gas	Air	Air
Driving gas pressure (MPa)	0.7	0.7
Driving gas temperature (°C)	25	80
Powder feed rate (g/s)	0.2	0.1
Nozzle transverse speed (mm/s)	75	50, 100, 200
Spray distance (mm)	10	10, 30, 50
Number of spray pass	1	1, 3, 5

Figure 3a–c show the effect of spray distance (SD) and nozzle speed (NS) on the electrical resistance of the resulting surface. Higher SD generally resulted in greater electrical resistance, which can be attributed to the: (i) discontinuities on the surface: [25] and (ii) falling gas velocity at higher SD, resulting in lower deposition efficiency (DE) [41, 42]. The lowest electrical resistance was obtained at SD = 10 mm for all conditions. Notably, the electrical resistance decreased at larger spray passes (N ≥ 3). It is likely attributed to the further material consolidation of the as-deposited Sn layer under the high-speed impact of upcoming Cu particles. Moreover, the electrical resistance tends to increase at higher NS. The reason behind this is the short interaction time of the particles with the target surface, resulting in less-particles deposited.

Figure 3d–f show the effect of the CS settings on the resulting surface film thickness. Lower NS leads to relatively thicker metallization on the polymer surface. It can be attributed to the better focusing of the sprayed particles onto a certain area at lower NS [43]. Conversely, when the NS is increased, the particles experience a shorter interaction time with the substrate, resulting in a thinner metal film on the target substrate per unit of time [44]. This phenomenon also explains the higher electrical resistance at increased NS (see Figs. 3a–c). Overall, the minimum film thickness of 39 µm occurred at NS = 200, SD = 50, and N = 3 while the maximum thickness of 58 µm was obtained at NS = 50, SD = 30, and N = 1. Remarkably, no significant increase in film thickness at larger spray passes (N > 1) was observed for Cu over-coating on the as-Sn deposited surface. This phenomenon is likely due to potential erosions on the polymer surface caused by the high-speed impingement of hard Cu particles onto the pre-metallized polymer surface [28]. The upcoming Cu particles likely tend to rebound from the as-metallized surface due to the metal–metal interaction rather than polymer-metal impact, thereby resulting in a lower deposition efficiency (DE). Gillet et al. demonstrated that the application of a two-layer (N = 2) CS coating using Cu particles resulted in a reduction of DE from 28.7% (first layer) to 15.4% (second layer) [16]. Furthermore, an increase in the number of CS passes could lead to a widening of the spray width, resulting in a higher occurrence of pores and cracks in the outer layer of the coating [45, 46]. Collectively, the optimal CS process settings were identified at SD = 10 mm, NS = 200 mm/s, and N = 1 pass, considering the resulting coating's low electrical resistance, minimal standard deviation, moderate coating thickness, and efficient processing speed.

3.2 Microstructure

SEM characterizations in Figs. 4a–d were carried out to evaluate the microstructure of the resultant coatings. As

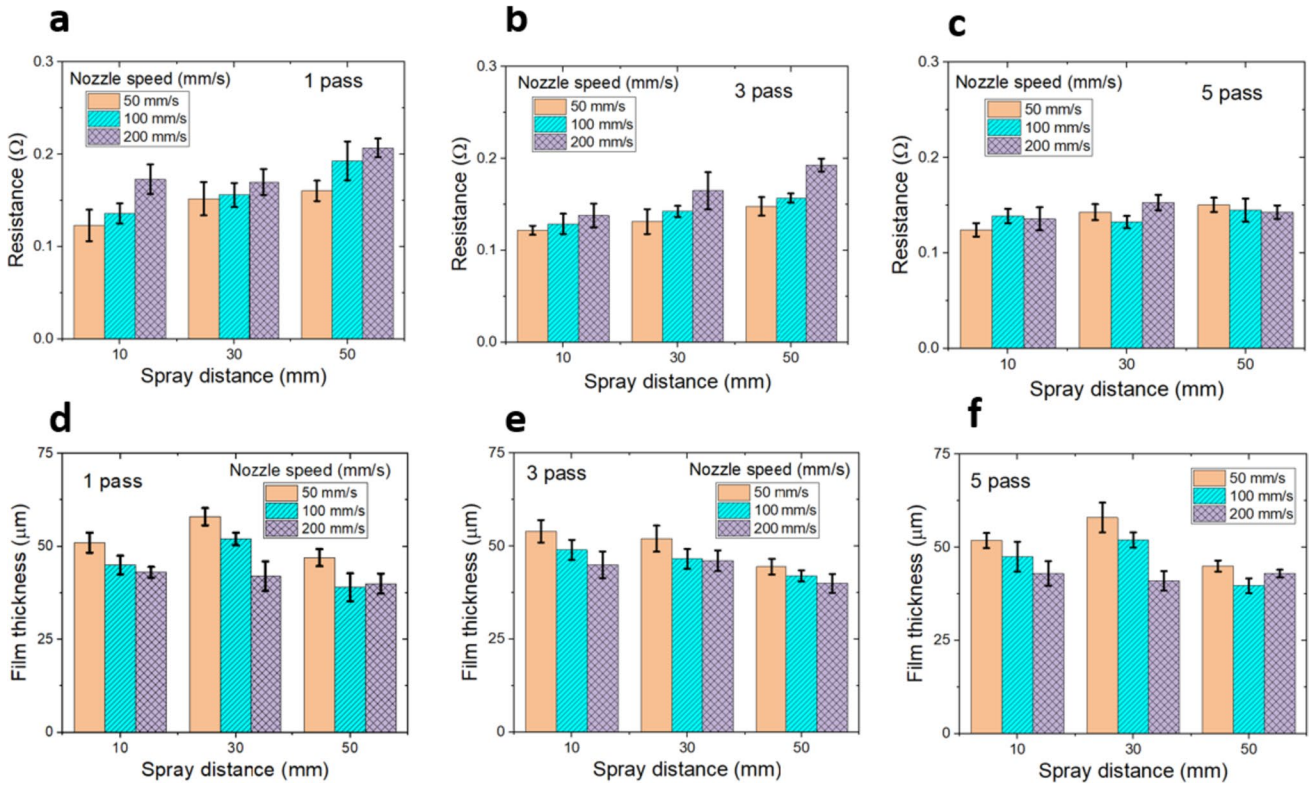


Fig. 3 Effect of nozzle speed, spray distance, and the number of spray passes on the electrical resistance and surface film thickness

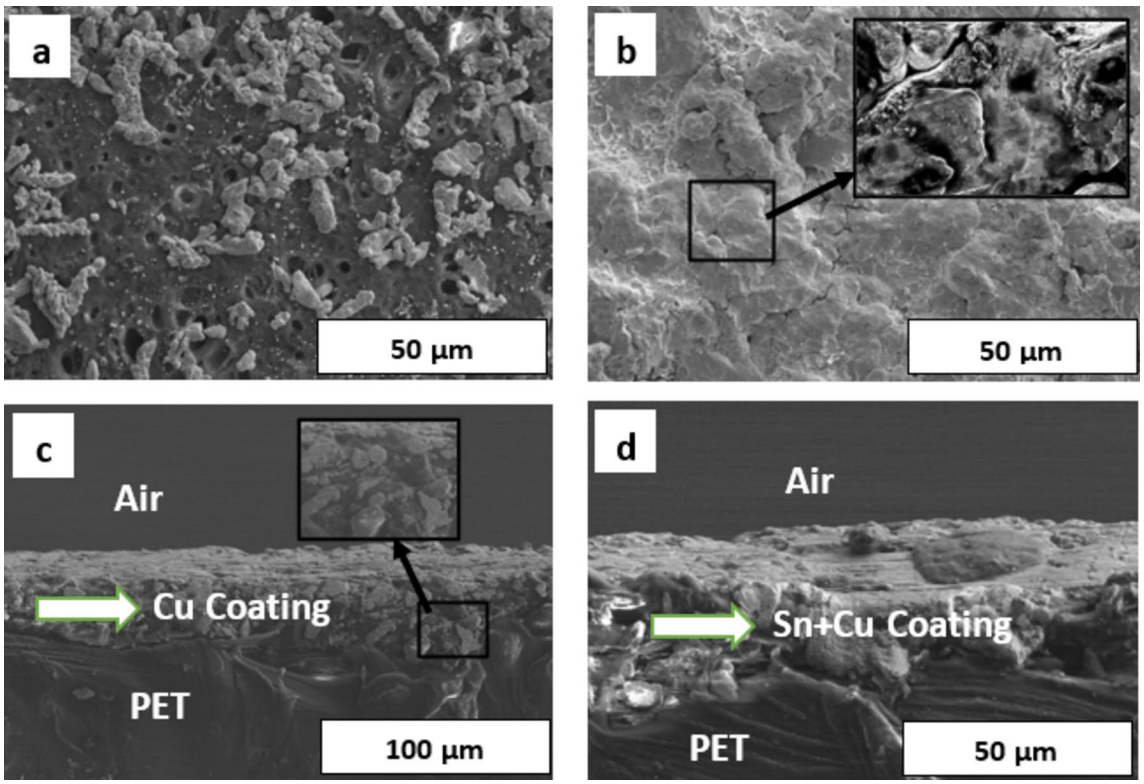


Fig. 4 Surface SEM images of **a** Cu and **b** hybrid (Sn + Cu) surface; cross-sectional SEM images of the **c** Cu, and **d** hybrid surface

seen in Fig. 4a, no continuous Cu deposition was achieved on the bare PET surface without Sn interlayer. Instead of deposition, Cu particles severely eroded the polymer surface, resulting in high porosity (see Fig. 4a). The authors also experienced the same phenomenon when Cu feedstock was cold sprayed on other thermoplastics such as ABS and Nylon 6 [8, 21]. The reason lies in the localized erosions on the polymer surface due to the high-speed impingement of hard Cu particles onto the soft thermoplastic polymers, resulting in no homogenously conductive coating [7]. The cross-section microstructure in Fig. 4c also confirmed the existence of pores along the interface.

On the other hand, as seen in Fig. 4b, the Sn interlayer facilitated the bonding of the Cu particles, resulting in a dense hybrid (Sn + Cu) metal surface on the PET surface. The cross-section SEM analysis in Fig. 4d indicates that Cu particles were successfully bonded on the pre-deposited Sn interface, producing a hybrid surface. Taken together, the Sn deposit effectively served as an inter-layer to facilitate the Cu over-deposition, which led to a hybrid (Sn + Cu) surface on the flexible polymer substrate.

EDX results of the resulting hybrid surface are also given in Fig. 5 in terms of atomic percentage (at %) (top panel) and elemental distribution map (bottom panel). The main elements of the resulting hybrid surface are Cu, Sn, C, and O, having atomic ratios (at%) of 76.7% Cu, 10.3% Sn, 7.1% C, and 5.9% O, respectively. As such, the EDX results suggest that the PET surface was successfully metallized with Cu and Sn particles. It can also be seen that the fabricated hybrid surface has a relative amount of oxide content (5.9%), which is likely attributed to the formation of the oxide layer on the resultant coating.

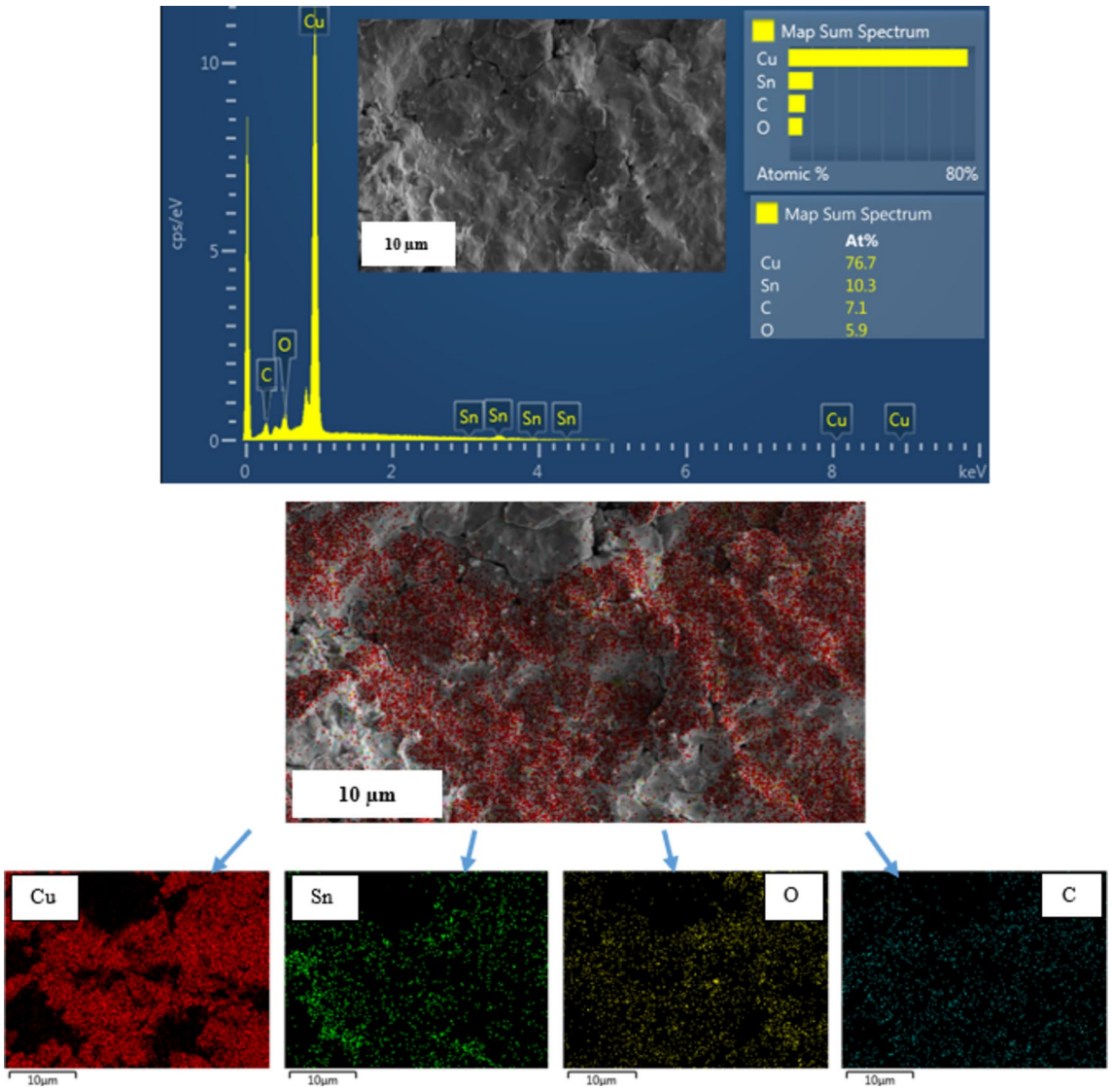


Fig. 5 EDX analysis of the fabricated hybrid (Sn + Cu) surface

3.3 Electrical Conductivity and Adhesion Strength

The electrical resistivity of the resultant hybrid surface was calculated using Eq. 1 where 4.532 is the correction factor, Rs is the average sheet resistance (Ω/sq), and t is the thickness (μm) of the metal film [47]. The average sheet resistance (Rs) of the resulting coating was measured as 0.0074 Ω/sq using a 4-point probe system at a constant current of 100 mA. Hence, the conductivity (i.e., inverse of resistivity) of the fabricated hybrid surface was calculated as 5.96×105 $S.m^{-1}$. This indicates promising electrical conductivity to utilize the fabricated multifunctional hybrid surface in numerous flexible electronics applications.

$$\sigma = \left[4.532 \times R_s \times t\right]^{-1} \quad (1)$$

We also tested the electrical stability of the resultant surface under various deformations such as bending and twisting. For this purpose, a red color light-emitting diode (LED) light circuit was designed on the polymer to test the electrical conductivity and adhesive strength of the surface under mechanical deformations. As seen in Fig. 6a, the circuit remained highly conductive under severe bending and twisting deformations, indicating high adhesive strength and electrical stability. Also, as given in Fig. 6b, we analyzed the relative resistance (R/Ro) variation of the electrodes for 30 days of storage at ambient conditions. The resistance did not increase more than 20% over a 1-month storage, proving the electrical stability of the resultant hybrid surface.

Lastly, we characterized the adhesion strength of the surface according to the ASTM D3359 cross-cut tape test method [38], and the results were evaluated based on the ASTM tape test scale. According to the cross-cut tape test, the surface achieved a score of 5B level, having less than 5% removed area on the surface (see Fig. 6c), indicating a strong interfacial adhesion strength.

3.4 Wettability Evaluation

The wettability of the surface is evaluated by water contact angle (CA) data for various surface configurations such as: (1) bare PET; (2) Sn surface; and (3) hybrid (Sn + Cu)

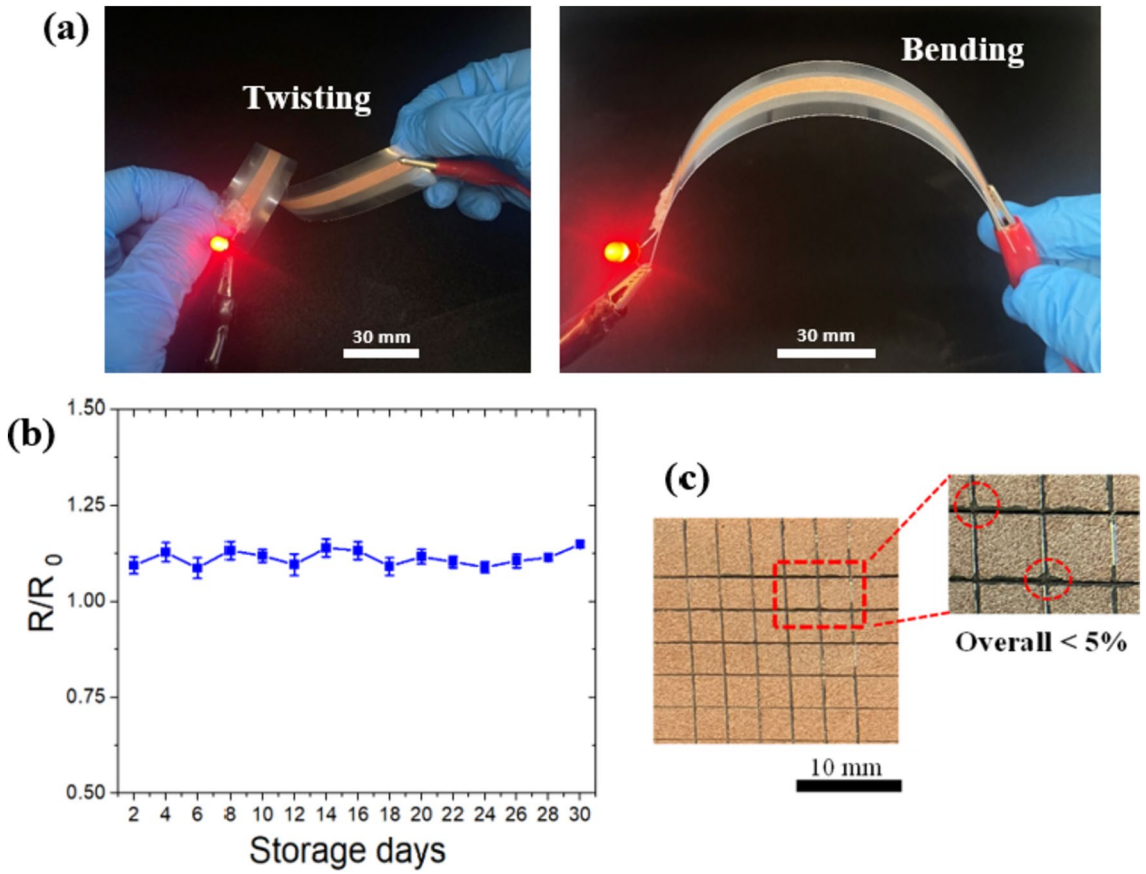


Fig. 6 **a** Testing the electrical stability of the fabricated hybrid surface under different mechanical deformation such as twisting (left panel) and bending (right panel), **b** the relative resistance (R/R0) change of the hybrid surface over storage days; (Electrode length = 20 mm and width = 5 mm), and **c** cross-cut adhesion test results

surface. As seen in Fig. 7a–c, the CA of bare PET, Sn, and hybrid surfaces were recorded as 70°, 85°, and 122°, respectively. As compared to solely Sn-coated polymer, the hybrid surface turns out to be hydrophobic (contact angle > 90°). The improved hydrophobicity of the surface can be attributed to: (i) reduced surface energy due to the dendritic-shape morphology of the Cu particles [48–50] and (ii) the micro-roughness of the resultant surface [50]. The results are comparable with the literature, in which Cu deposits on metals, glasses, and ceramics improved surface hydrophobicity [51–53].

We also investigated the effect of the Cu over-coating on the surface wettability under various Cu weight ratios, ranging from 15 to 70%. As seen in Fig. 8, an increase in the Cu content up to 70 wt% led to a hydrophobic surface structure, increasing the CA from 85° to 130°. The results suggest that the hybrid surface significantly improved the hydrophobicity of the surface, thereby having the potential for the applications where surface hydrophobicity is desired such as antibacterial and even self-cleaning surface applications.

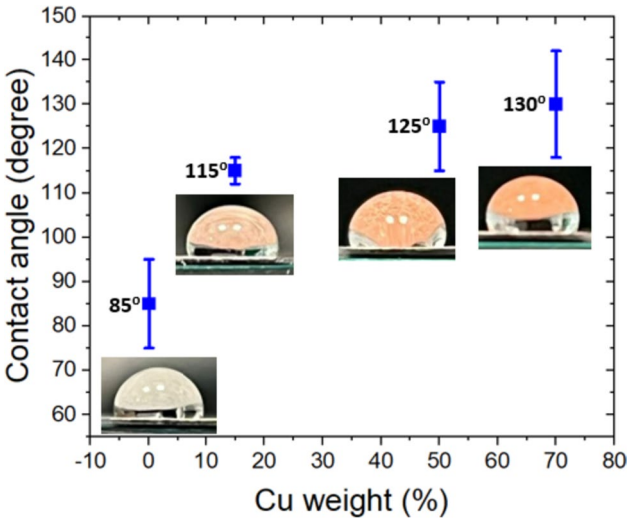
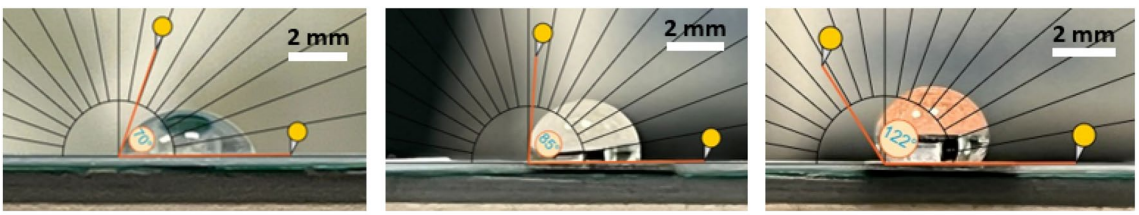


Fig. 8 Contact angle variation of the surface with various Cu weight ratios

3.5 Antibacterial Performance

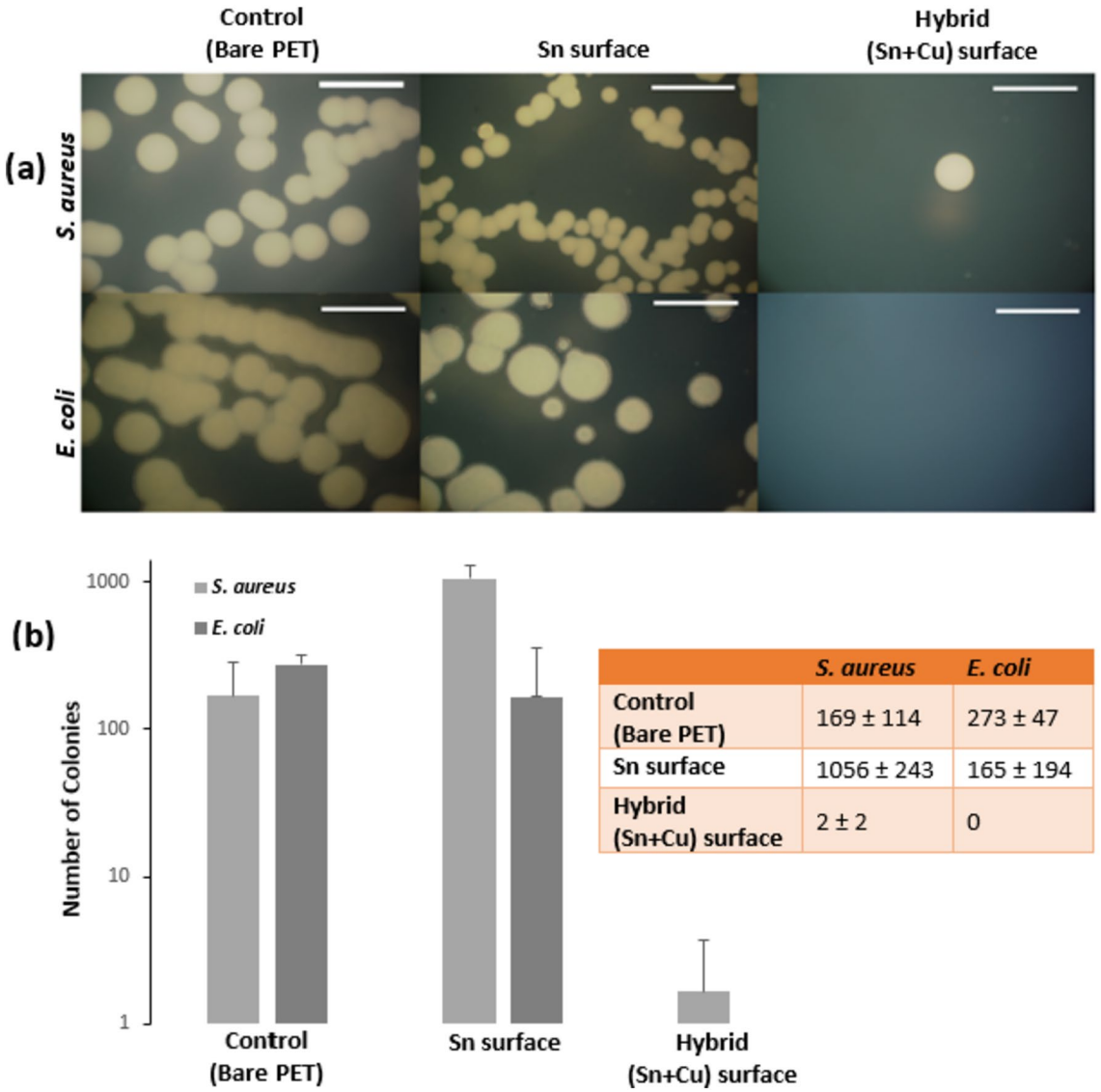
Antibacterial performance of the fabricated hybrid surface was evaluated by examining the inhibition of bacterial growth. The antibacterial activity of the surfaces was tested using Escherichia coli (*E. Coli*) and Staphylococcus aureus (*S. aureus*) bacteria, which are widely used in the antibacterial surface tests [54–56]. Details of the antibacterial test procedure are provided in Appendix A2.

The antibacterial performance of various surfaces is presented in Fig. 9a, b. As seen from the portions of the agar plates after bacteria culture in Fig. 9a, agglomerated groupings of bacteria for the control (bare PET) and Sn surfaces were observed, demonstrating that most of the streaked bacteria were still viable. Conversely, the handful of viable colonies produced by *S. aureus* and these colonies were inhibited by the hybrid surface (see Fig. 9b). Despite the bacteria growing on both the control group and on the Sn surface, only a negligible amount of bacteria strain survived under the 24 h of exposure to the hybrid surface. In the case of *S. aureus,* only a handful of bacteria survived to be able to produce colonies and for *E. coli* none of the bacteria were still viable after 24 h. The antibacterial test results proved that > 99% of the bacteria were extinguished by the fabricated hybrid surface after 24 h of exposure. The disappearance of bacterial colonies shows that the developed hybrid surface demonstrates excellent antibacterial performance. As such, the *in-vitro* antibacterial test revealed the promising antibacterial performance of the multifunctional hybrid surface that resisted the colonization of bacteria as it inhibits > 99% of the bacteria colony.



Contact angle (deg.)	PET	Sn coated surface	Hybrid (Sn+Cu) surface
	70°±15	85°±10	122°±10

Fig. 7 Water contact angle of the **a** bare, **b** Sn, **c** hybrid (Sn + Cu) surface, (Cu weight ratio is 70 wt% for the hybrid surface)



	S. aureus	*E. coli*
Control (Bare PET)	169 ± 114	273 ± 47
Sn surface	1056 ± 243	165 ± 194
Hybrid (Sn+Cu) surface	2 ± 2	0

Fig. 9 Antibacterial characterization of the surfaces against *S.aureus* (top panel) and *E. coli* (bottom panel) **a** Bacteria culture on the surfaces after 24 h of incubation (scale bar = 2 mm), **b** Bacterial growth on the surfaces (number of experimental groups, n = 3)

4 Conclusion

In this study, highly flexible, conductive, and antibacterial surfaces were fabricated by the CS technique. First, the Sn particles were deposited on the flexible polymer (PET) substrate to achieve an electrically conductive surface. Next, the subsequent over-Cu deposition ensured the hydrophobic and antibacterial surface. Through this manufacturing approach, a multifunctional (flexible + conductive + antibacterial) surface was achieved at low-processing temperatures without a need of heat, vacuum, and precursor. The following results can be drawn from the present work:

- The resulting hybrid surface showed promising electrical conductivity (5.96×10^5 S.m^{-1}), flexibility, stability, and adhesive strength under various mechanical deformations such as bending and twisting without compromising electrical conductivity and structural integrity.
- Cu over-coating significantly increased the water CA (from 85° (Sn) to 122° (Sn + Cu)), ensuring hydrophobic surface characteristics.
- An increase in the Cu weight ratio led to a higher water CA (1 30°), which enhanced the surface hydrophobicity.
- *In-vitro* antibacterial tests revealed that the fabricated hybrid surface can inhibit > 99% of bacteria.

Taken together, the findings demonstrate the promising potential of the fabricated multifunctional hybrid surfaces for various application domains, including printed flexible electronics, smart thin films, polymer sensors, hybrid electronics, and many more. Furthermore, they can particularly serve as effective anti-bacterial surfaces, efficiently deactivating the bacteria on high-touch areas with conformal geometry. Moreover, in consumer electronics, they could facilitate the development of innovative electronic gadgets with heightened functionality and performance. Therefore, the multifaceted capabilities of these surfaces in this study have the potential to drive advancements in multifunctional flexible electronics and beyond.

Appendix

Full Factorial Design of Experiments for the CS

See Table 2.

Antibacterial Test Procedure

The antibacterial testing was modified from the standard methods of the International Organization for Standardization [ISO 221961; 'Plastics – Measurement of antibacterial activity on plastic surfaces']. Six specimens (50 cm × 50 cm) with bare PET and Sn-surfaces were considered as the untreated and bacterial viability control groups. In addition, three samples of the same dimensions for hybrid (Sn + Cu) surface were prepared as the treatment group. *E. coli* was grown in lysogeny broth while *S. aureus* was grown in tryptic soy broth. Bacteria were grown to an optical density between 0.23 and 0.25 when measured at 600 nm with a UV spectrometer. Then 0.4 ml of bacteria was placed on the surface of each sample and a piece of 22 × 40 mm^2 microscope cover glass (Fisher, 12-543A) was placed on top. The treated and untreated samples were then placed in the incubator at 35 °C for 24 h. Fluid with bacteria was then removed from the surface by rinsing gently with 10 ml of 1 × Phosphate-Buffered Saline (PBS). This was then diluted to tenfold with 1 × PBS and placed on an agar cell culture plate by streaking. Plates were incubated at 35 °C for 24 h after which the number of colonies was counted. The samples for measuring the bacteria viability on the untreated surface were removed from the surface using 1 × PBS and cultured on agar plates using the exact method mentioned above however without the 24-h incubation on the sample surface.

Table 2 Full-factorial experimental design layout

Run	Factors			Results	
	Nozzle speed (m/s)	Spray distance (mm)	Number of spray pass	Average film thickness (μm)	Average resistance (Ω)
1	50	10	1	51	0.123
2	50	10	3	54	0.122
3	50	10	5	51.8	0.124
4	50	30	1	58	0.152
5	50	30	3	52	0.131
6	50	30	5	58	0.143
7	50	50	1	47	0.160
8	50	50	3	44.5	0.148
9	50	50	5	45	0.150
10	100	10	1	45	0.136
11	100	10	3	49	0.129
12	100	10	5	47.5	0.139
13	100	30	1	52	0.156
14	100	30	3	46.6	0.132
15	100	30	5	52	0.147
16	100	50	1	47	0.193
17	100	50	3	44.5	0.157
18	100	50	5	45	0.145
19	200	10	1	43	0.173
20	200	10	3	45	0.138
21	200	10	5	43	0.136
22	200	30	1	42	0.170
23	200	30	3	46.1	0.165
24	200	30	5	41	0.153
25	200	50	1	40	0.207
26	200	50	3	41	0.193
27	200	50	5	43	0.143

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Declarations

Conflict of Interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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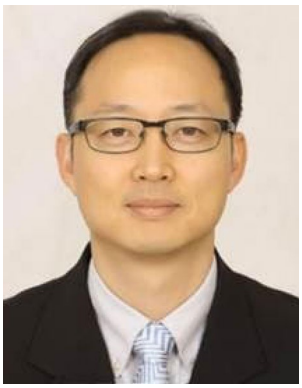

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