




Research Article

Design and Fabrication of Metallic-Conductive Polymer-Based Hybrid Film Interconnections for Stretchable Electronic Devices

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Stretchable circuit is a technological innovation that has transformed the microelectronic landscape due to its enormous applications in the field of medicine. The consistency or durability of health monitoring devices can increase the dependability with which non-invasive clinical measures are collected. Metal-conductive polymer (CP) hybrid interconnects and metal-polyimide dual-layered interconnects were all produced as stretchable interconnections. Stretchable substrate for all of the interconnects was selected as soft elastomer polydimethylsiloxane (PDMS). However, the PDMS substrate presents challenges because it is temperature sensitive, limiting the process temperature. The extreme hydrophobic nature of the PDMS surface makes it difficult to deposit components that contain water and results in poor adhesion with different metals. Following the development of processes for fabricating materials on the PDMS substrate, methods for resolving these issues were investigated.

1. Introduction

Polydimethylsiloxane (PDMS) is a polymer made of SiO (CH₃)₂ repeating siloxanes, as a stretchy substrate, as shown in Figure 1. The hydrogen bond formed between both the silanol group on the silica surface and oxygen in PDMS gives it a high adherence to silica [1]. A PDMS solution is spun-coated at 500 rpm for seconds on top of Si to achieve sufficient thickness. It is then dried for 2 hours at 90°C. Before the metal was evaporated with an e-beam, an oxygen plasma treatment was conducted for a brief period. Gold (Au) is ductile, heavily conductive, and insoluble to oxidation, it was selected as the metallic film in this case. To improve adhesion even further, a thin layer of Cr or Ti, i.e., approximately 5 nm, may be evaporated well before Au. Wet etching and photolithography were used to pattern the metallic thin film (MTF). The hydrophobic

properties of PDMS substrate are one of the fabrication process difficulties. This PDMS characteristic causes the deposited conductor to adhere poorly. According to Figure 1, the oxygen plasma continues to change the layer of the PDMS substrate by oxidizing CH₃ in the outer chains, and thus changing the PDMS surface from hydrophobic to hydrophilic [2].

Next-generation flexible transparent devices using poly(3,4-ethylenedioxythiophene) (PEDOT): polystyrene sulfonate (PSS) conductive films were developed. However, as a result of defect formation or incomplete layer, electrodes exhibit extremely high mechanical deformation and low electrical conductivity. This is a significant and appealing Conductive Polymer (CP) with unique properties such as outstanding solution fabrication ability and dispersibility, extremely good chemical and electrochemical stability, high and controllable conductivity, good optical transparency, and biocompatibility.

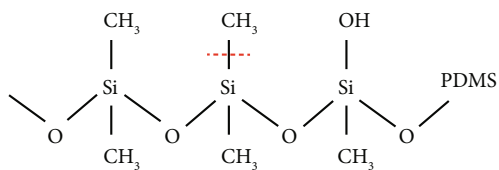


FIGURE 1: OH-bond to transform PDMS surface.

It is available commercially in organic electronics, including electronic chemosensors.

Annona reticulata dry leaf is used to make negative charge Au and Ag nanoparticles, which are then used to make TF of PEDOT:PSS nanomaterials. The layout and morphological characteristics of nanocomposite vary significantly with respect to the type and concentration of nanoparticles. The complicated PEDOT:PSS CP is the most investigated CP for textile applications. Since this is widely available in dispersion version, which is compatible with existing textile processing applications. Organic Electrochemical Transistors (OECTs) are an adaptable category of biosensing elements because of their sensitivity, potential cost, and system integration benefits. The electrical properties of this electrode are said to be heavily influenced by the film's construction. Out-of-plane longitudinal detachment between adjoining PEDOT domains and mutational reinterpretations in-plane process segregation may occur due to film processing. The operation of OECTs is thickness-dependent, which is explained as how the thickness of the film promotes microstructure tuning all through long-lasting thermal annealing (LTA) in controlled atmosphere. The creation of a PEDOT:PSS polymeric film for use in flat panel displays, control boards, and other optoelectronic is a significant advancement.

New composite materials that display high electrical conductivity and mechanical softness still encounter challenges in terms of chemical degradability, stability, and compatibility with current Micro-electro-mechanical System (MEMS)/ Complementary Metal-oxide-semiconductor (CMOS) technology. Through a variety of methods, this research offers solutions to these problems. Composites are the well-liked choice for stretchable interconnects. It displays greater stretchability and higher electrical conductivity. Their high contact resistance due to the dielectric polymer matrix is one of the disadvantages. The proposed composite includes PEDOT:PSS as the polymer matrix and graphite flakes as the conductive filler to reduce contact resistance and sheet resistance. This composite can provide better stretchability compared to carbon-based composites.

2. Related Works

By integrating polyvinylcycamide with PEDOT:PSS, Ma et al. [3] reported an intrinsically flexible conductive composite film with high electrical properties and minimal damage in challenging environments. It has flexibility properties and substantially improved electrical conductivity. A wearable touch panel device has been fully integrated with the composite film. Gao et al. [4] described the use of PED-

OT:PSS and its composite materials in sensors and electrochemical, which detect the presence of gaseous chemical analytes or liquid-phase analytes such as inorganic compounds ions, pH, ammonia (NH_3), hydrogen peroxide, CO_2 , CO, NO_2 , and organic solvent vapours such as acetone, methanol, and others. The optimization of PEDOT structural, PSS architectural, and morphological properties, as well as composites with other additives, was extensively discussed.

According to Sarkar et al. [5], the examination of shape and film thickness with infrared can demonstrate the films' composite composition. With an increase in nanoparticle concentration, the composite films' in-plane DC conductivity is moderately improved. Electrical responses based on impedance spectroscopic analysis depend on nanoparticle absorption. Structure changes take place after the composite is created. Tseghai et al. [6] developed a method on PEDOT:PSS-based textiles semiconductor, their techniques of implementation in textiles, and their own applications. One method for increasing the mechanical adaptability of CPs is to create a nanoparticle out of common resource polymers with high versatility and stretchability, such as polyurethane. These conductive fibres have been created utilizing solution spinning or electrospinning techniques. And this can be applied to cloth via coating/dyeing, printing, and monomer polymerization. Conductive textiles based on this criterion have been used to make actuators, sensors, interconnections, antennas, storage devices, and energy harvesting devices.

Ji et al. [7] described an innovative organic-inorganic material made of phosphomolybdic acid and PEDOT:PSS as having good solvent corrosion resistance and great hydrophilicity with the printed top Ag nanowires and active layer. Fully coated translucent tools with spray-coated Ag nanowire top electrodes and doctor-blade-coated layers outperformed cells with a thermally evaporated MoO_3 layer in terms of overall performance and Average Visible-light Transmittance (AVT). The highest Power Conversion Efficiency (PCE) was 5.01%, with an outstanding average visible light transmittance of 50.3% (with a PCE of 5.77% and AVT of 19.5%). D'Angelo et al. [8] compared the response of OECTs to LTA with similar channel thickness that had undertaken a quick annealing. This process increased the amplification capacity of OECTs, as demonstrated. The LTA procedure resulted in firmer charge carrier modulation on thinner layers and combined solution processing ability to manipulate the microstructure with the impact of post-deposition processing.

Kleber et al. [9] presented a new transparent hydrogel system composed of the synthesised hydrogel and the CP:PEDOT as a protective coating for brain interfaces. The composite material has the ability to form an interpenetrating network and to be linked covalently with the surface electrodes. It can also be photolithographically shaped just to affect specific electrode places. With the help of the disclosed material, it is feasible to modify the surface of neural probes, combining the advantageous characteristics of conducting polymer hydrogel (CPH) with the superior qualities required for higher quality brain microelectrodes. The effort of Zabihi and Eslamian [10]

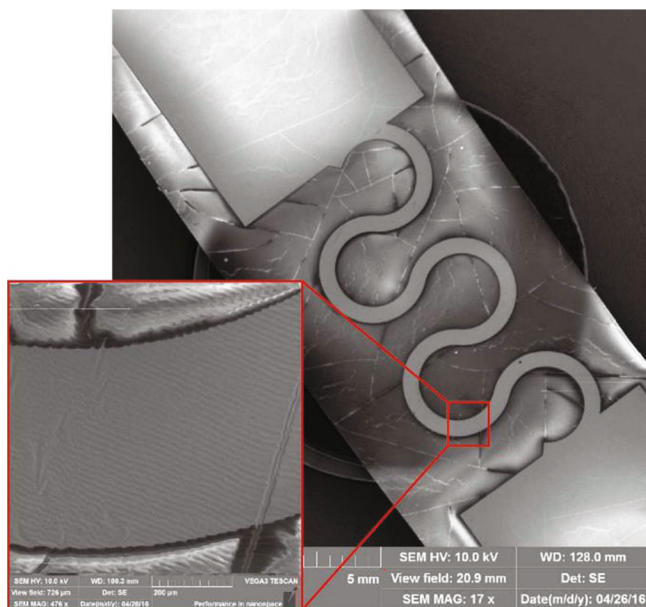


FIGURE 2: A SEM of a patterned Au interconnect on a PDMS substrate.

to substitute the expensive and restrictive conductive oxides used for the electrodes in Thin Film Diode (TFDs) is demonstrated. The spray-on transparent graphene electrode (TGE) have surface and film coverage and morphological characteristics that are comparable to or superior to PEDOT. A perovskite solar cell with a PCE of 3.54% was used as a proof-of-concept. As contact electrodes for printed organic field effect transistors, Sanyoto et al. [11] used dimethyl sulfoxide (dms) as a dopant, and the optimum field effect accessibility for diketopyrrolopyrrole-thieno[3,2-b]thiophene organic field effect transistors (DPPT-TT OFET) was $0.49 \times 0.03 \text{ cm}^2 \text{ V}$.

Kraft et al. [18] proposed a conducting polymer-based ink for flexible interconnections. Wide area handling is flexible, non-invasive, and maskless due to the tunability of inkjet printing paradigm. The imprinted PEDOT:PSS-based interconnections can withstand stresses of more than 100% and have conductivity in the range of 700 S cm^{-1} . Iron (Fe) nanoparticles were used by Zhang et al. [19] as the conducting polymer and polycaprolactone (PCL) as the insulation material for the creation of degradable conducting polymer coatings that made up the interconnects. Under varied degrading settings, the electrical characteristics of the composite were examined. At 17% Fe concentration, electric percolation was seen, while larger content fractions showed more consistent resistance during the duration of physical deterioration. The implementation of chip interconnect by utilizing polymer-based optical waveguides was introduced by Nieweglowski et al. [20]. They offered polymer-based methods for optical connection among waveguides and electronic devices. This strategy relied on bendable substrate with integrated rectangular polymeric optical waveguides. These are investigated as a direct incorporation of polymer waveguides utilizing the dicing technique.

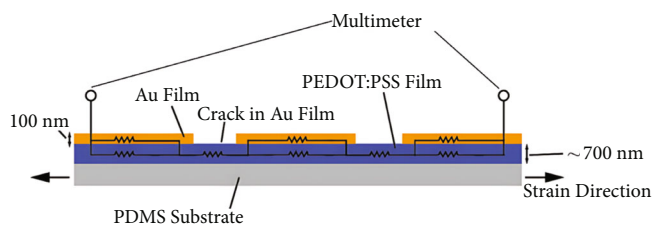


FIGURE 3: Schematic cross-section view of Au-PEDOT:PSS.

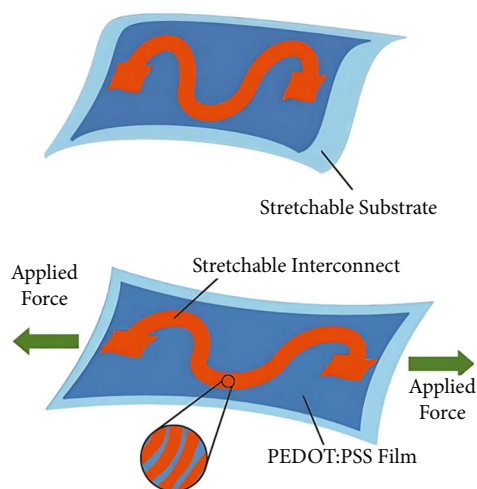


FIGURE 4: Schematic of strain applied on interconnect with PEDOT:PSS.

3. Materials and Methods

The significant interfacial stress was created between the PDMS substrate and the deposited metallic layer. The metallic coating develops an intrinsic stress during the metal deposition process. This stress tries to release after the film is deposited. The metallic sheet releases the stress since PDMS is a soft substrate, which causes cracks. Deposition rate (DR) is defined as the increment in multi-film thickness (MFT) with respect to the fabrication time. As per the experimental analysis, lower metal DR of 0.4 nm/s can help to reduce internal stress [12]. The DR was therefore marked, and the Au deposited has a thickness of 50 nm . When viewed under a microscope, the density of fissures created within the formed layer dramatically decreased. Plasma treatment based on oxygen over PDMS was examined for varying lengths of time to further enhance the procedure. To protect the moisture, etch of the Au layer, a photoresist surface was used. After a 15-second soak in potassium iodide (KI) solution, the Au was wet inked. A scanning electron microscope (SEM) image obtained using ZEISS GeminiSEM 560 is used to examine the patterned connections as illustrated in Figure 2. Significant folds on the deposited PDMS substrate and Au film can be seen in the highly enlarged SEM image. This surface shape could be attributed to the PDMS substrate's elasticity, which resulted the surface to contract and expand during the metal deposition process [13].

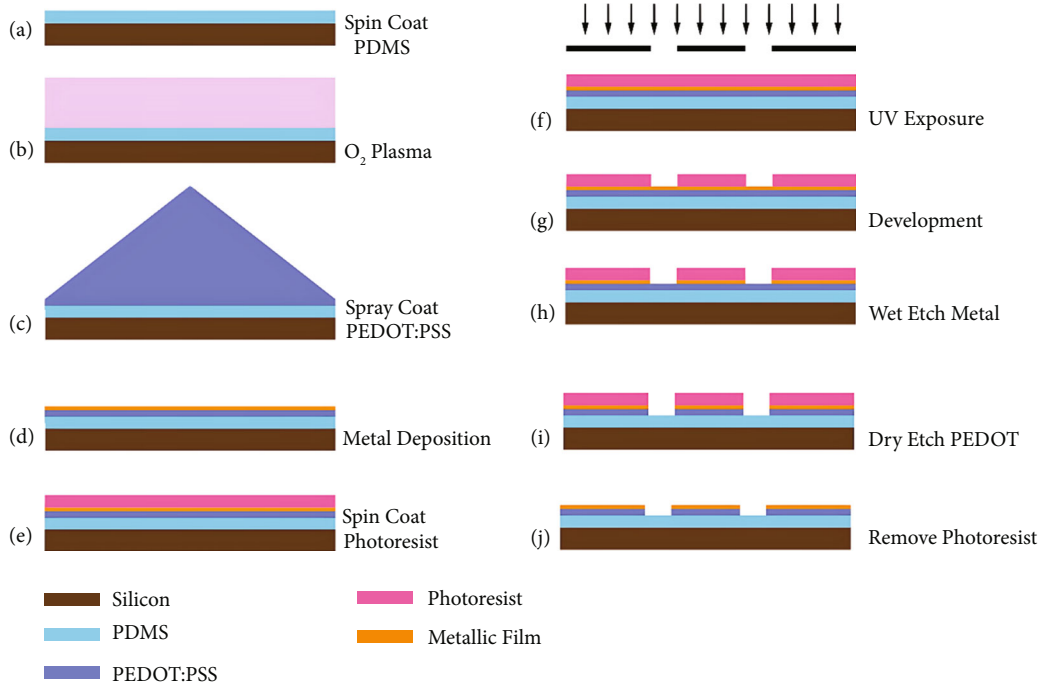


FIGURE 5: Flow of fabrication process involving spray-coated PEDOT:PSS.

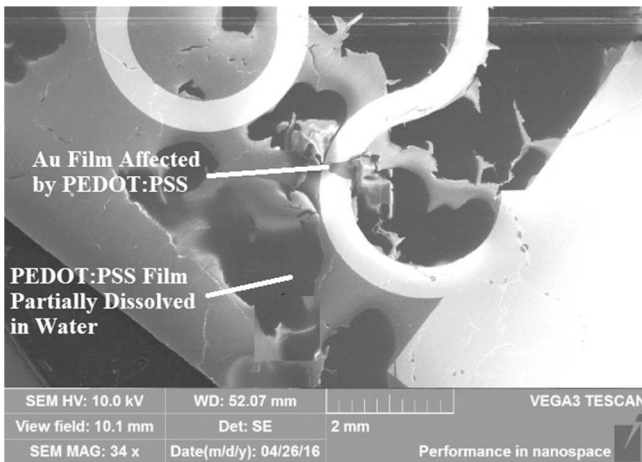


FIGURE 6: SEM image of fabricated IC with Au film in PEDOT:PSS.

TABLE 1: Changes in morphology of Ti-Au films under different conditions.

Metal process	Condition	Roughness
Ti-Au (10–50 nm)	O ₂ plasma 10 s at 80 W	Rq: 17.3 nm Ra: 14.4 nm
Ti-Au (10–50 nm)	O ₂ plasma 30 s at 80 W	Rq: 10.7 nm Ra: 8.9 nm
Ti-Au (10–50 nm)	O ₂ plasma 60 s at 80 W	Rq: 2.21 nm Ra: 1.75 nm

3.1. Metallic-Conductive Polymer Hybrid Film Interconnects. Soft conductors have been researched as a potential solution to the metallic conductor cracking problem. This film has a

moderate electrical conductivity when compared to metallic conductors. Such CPs are also moisture-sensitive, which makes long-term stability difficult. A hybrid construction with metal-CP interconnects was suggested to solve these problems. The metal can perform as a barrier against contaminants as well as enhance the hybrid structure's electrical conductivity. External strain-induced cracks in the top MTF, as shown in Figures 3 and 4, will not surely result in an interruption in the electrical connection. The metallic islands continue to form the electrical conduct path, but they are only loosely connected. HTC Instruments DM-15S TRMS multimeter is used to measure conductivity and resistance.

A conductive polyelectrolyte complex is the PEDOT:PSS. Positively charged PEDOT serves as the conductive component, and the PSS component balances the doping charges and facilitates PEDOT's water dispersion [14]. PSS is good in preventing conductive PEDOT molecules from connecting with one another and reducing the electrical conductivity [15, 16]. The schematic representation of interconnect on a polymer base while applying strain is depicted in Figure 4. Here force is applied from both ends, and the morphology of the interconnect changes without affecting the electrical properties.

3.2. PEDOT:PSS Spray Coating. Figure 5 illustrates the development of spray coating of PEDOT:PSS solution over a substrate of PD. The PEDOT:PSS solution was sprayed using compressed N₂ gas. 5% isopropanol (IPA) was mixed to make it easier to coat the PDMS substrate. 10 seconds of O₂ plasma treatment can significantly increase the angle of contact between the solution and the surface of PDMS. This makes it simpler to spread the PEDOT:PSS over the PDMS substrate. You can run spray coating multiple times to change the PEDOT:PSS thickness of film. Based on the

TABLE 2: Comparison of morphology change of Au under various plasma conditions.

Metal (50 nm)	Process	Duration	AFM surface	Roughness
Au	No plasma	Nil	Rq: 11.6 nm Ra: 9.5 nm	Rq: 7.65 nm Ra: 3.02 nm
Au	O ₂ plasma	10 seconds	Rq: 10.7 nm Ra: 8.9 nm	Rq: 11.4 nm Ra: 10.11 nm
Au	O ₂ plasma	20 seconds	Rq: 2.21 nm Ra: 1.75 nm	Rq: 6.52 nm Ra: 6.12 nm
Au	O ₂ plasma	30 seconds	Rq: 1.42 nm Ra: 0.98 nm	Rq: 4.42 nm Ra: 6.78 nm

interferometer (Leitz Ergolux) measurement, each spray coating run produces a thick PEDOT:PSS film.

PEDOT:PSS film delaminates and dissolves, as seen in Figure 6. The top Ti–Au coating will be further pulled away from the substrate by the PEDOT:PSS film delamination. 50 nm thick MTF layer is deposited over the polymer layer. A possible solution to this problem is to switch from wet to dry etching of metal, depending on the clean room’s capabilities.

Due to its hydrophobicity, the purified PEDOT:PSS that was purchased is difficult to deposit directly on the PDMS substrate because it is dispersed in water. Despite giving the PDMS substrate enough oxygen plasma treatment, the solution has a tendency to separate from the substrate while spinning. Three low-boiling-point solutions were separately combined to help the solution spread uniformly. The PDMS substrates are initially placed on 1.5 cm × 2 cm square-shaped glass slides and exposed to plasma state O₃ for 60 seconds at an intensity of 80 W. Both Au and PEDOT:PSS films are put under a lot of strain during the peeling process, which causes the films to crack even before they are put to the test.

4. Results and Discussions

To further improve the process, an oxygen plasma treatment on PDMS was investigated under varied durations. A comparison between the plasma condition and substrate surface morphology of Ti–Au films has been investigated, as listed in Table 1. Confirmed by atomic force microscopy (AFM) (Nanoscope, Scan Asist Mode, Bruker Icon AFM, U.S) measurement, the surface roughness reduced from 17.3 to 10.7 nm after increasing the duration of oxygen plasma from 10 to 30 seconds. After 60 seconds of exposure, the surface becomes smooth, with a roughness of 2.21 nm. However, the cracks in the resulting film can still be noticed. Observed from an optical microscope, the density of cracks was reduced. The square root of the sum of the squares of the different heights and depths from the mean line (0.5) is defined as root mean square roughness (Rq). The surface’s average roughness is termed as Ra.

A further study was conducted to investigate the effect of depositing pure Au layer on top of PDMS. A comparison between the plasma condition and substrate surface morphology for Au films has been investigated, as listed in

Table 2. The surface roughness reduced from 11.4 to 6.52 nm after increasing the duration of oxygen plasma from 10 to 30 seconds. After 60 seconds of exposure, the surface becomes smooth, with a roughness of 4.42 nm. No cracks have been observed on all the samples in which the metal was deposited under different plasma conditions [22]. However, under the AFM scanning, the nano-scale cracks can be found. A short time (10 and 30 seconds) plasma does not benefit the Au film to significantly reduce the cracks. 60 seconds of oxygen plasma resulted in a crack-free metallic film on PDMS substrate.

The difference in thickness is visible for all of the samples. The sheet thickness increases with the increase in number of runs through which the polymer material is applied to the substrate. The thickness remains constant at 700 nm and the application process is stopped at that point. The variation in thickness with respect to number of runs is illustrated in Figure 7.

A sheet resistance measurement device is used to gauge the PEDOT:PSS film’s sheet resistance. By lengthening the spray runs, the measurement picks up a slight decrease in sheet resistance. The resistance varies between 3000 and 1000 Ohms. As the number of spray runs increases, the resistance decreases. This is an indication of the increment in conductivity of the polymer material used for interconnections. The variation in sheet resistance with respect to spray runs is illustrated in Figure 8.

After accounting for the thickness and sheet resistance measurements, it can be noted that the developed polymer material is well suited for the development of interconnects. Figure 9 shows the optical image of a sprayed 700 nm thick PEDOT:PSS film on a PDMS substrate obtained using Celestron TETRAVIEW 5MP digital microscope. It has a resistivity of about 0.0072 Ohm metre.

Figure 10 shows the fitted curve values vary greatly (standard error). This could be due to the probe’s shaky interaction with the specimen surface. Since the sample’s surface is soft, the metallic probes had no problems penetrating it [17]. The resistance of Pristine PEDOT:PSS varies from 1270 to 3300 Ohm, when the distance between the interconnects is varied from 10 to 30 mm.

Meanwhile, the AFM measurement was used to find morphology [21]. The doped-graphite powder effect increased the surface roughness. About 8.5 nm of depth and as a result is an increase in overall roughness from 3.84 to 6.31 nm. The electrical characteristics of composite were carried out with the help of an I–V characteristics probe station. The transfer line measurement (TLM) is used to measure the sheet resistance and contact resistance. As shown in Figure 10, proposed polymer-based interconnect provides better linear fitting for resistance respect to the varying distance when compared to un-doped PEDOT:PSS film [23].

The polymeric composite design benefits from both the fillers’ high conductivity and the polymers’ softness [24]. It displays a high elastic modulus, but the majority of polymers, which are also soft and supple like human skin which are insulating. When compared to its metallic equivalent, the polymer connector has less roughness. In comparison to metallic interconnects, such polymer composites

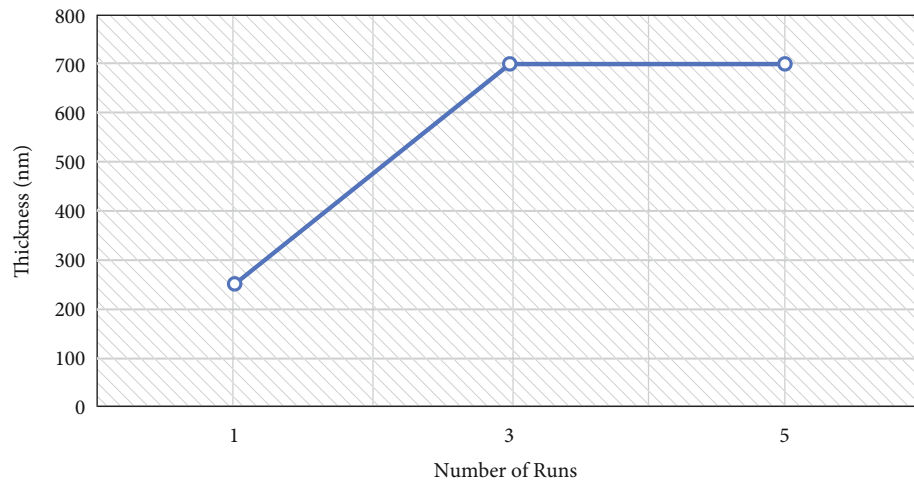


FIGURE 7: Thickness measurement.

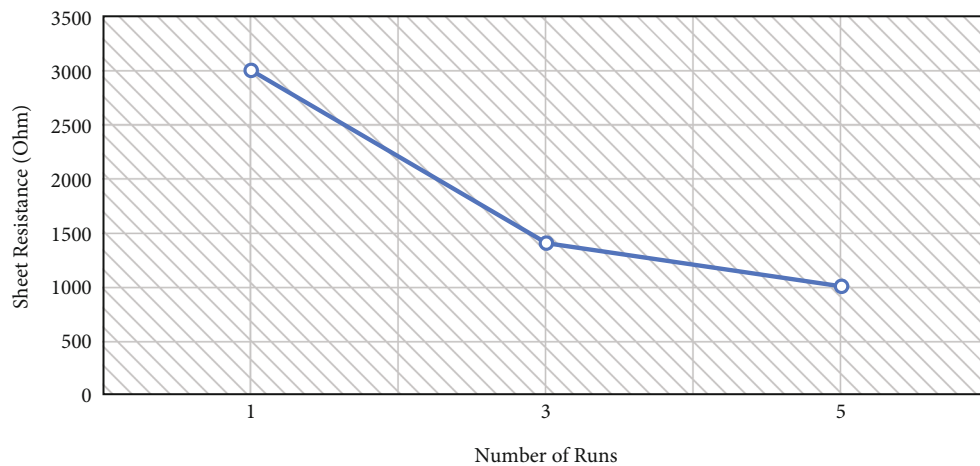


FIGURE 8: Sheet resistance measurement.

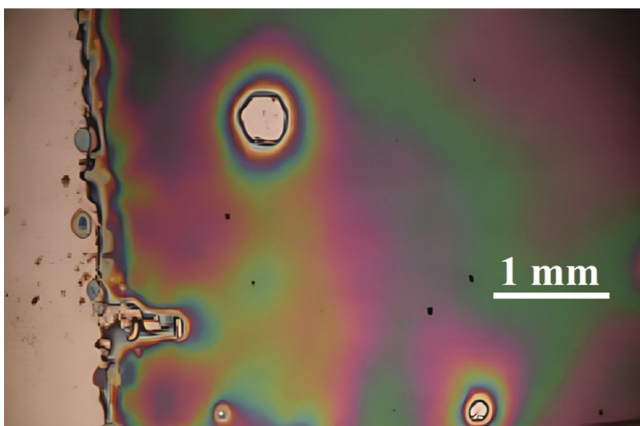


FIGURE 9: Optical image of PEDOT:PSS on PDMS substrate.

have higher stretchability values. As distance increases, the interconnection's resistance changes continuously [25]. Due to their availability, higher conductivity, and cost-effectiveness, polymers are frequently employed as conductive

fillers. However, in practice, bigger conductors are more expensive, and manufacturing technologies also have a limit on how thick they can be. The fabrication approach increases the polymer interconnects' durability while being very cost-effective.

5. Conclusion

This paper discussed the fabrication of polymer-based stretchable interconnects using various conductive materials. As a substrate for all types of interconnects, the elastomeric polymer PDMS was used. Au films have long been employed in conventional electronic devices as standard conductors. To deposit 50 nm MTF on soft substrates, an optimized fabrication process was developed. Microfabrication technology is used in the procedures. Resolution (up to 5 μ m), flexibility, and reproducibility are all advantages of these processes. Spin coating provides solution for uneven thickness on PDMS substrate. In order to create a hybrid conductive film, a second metallic layer was to be deposited on the PEDOT:PSS film. As a result, manufactured interconnect resolution falls short

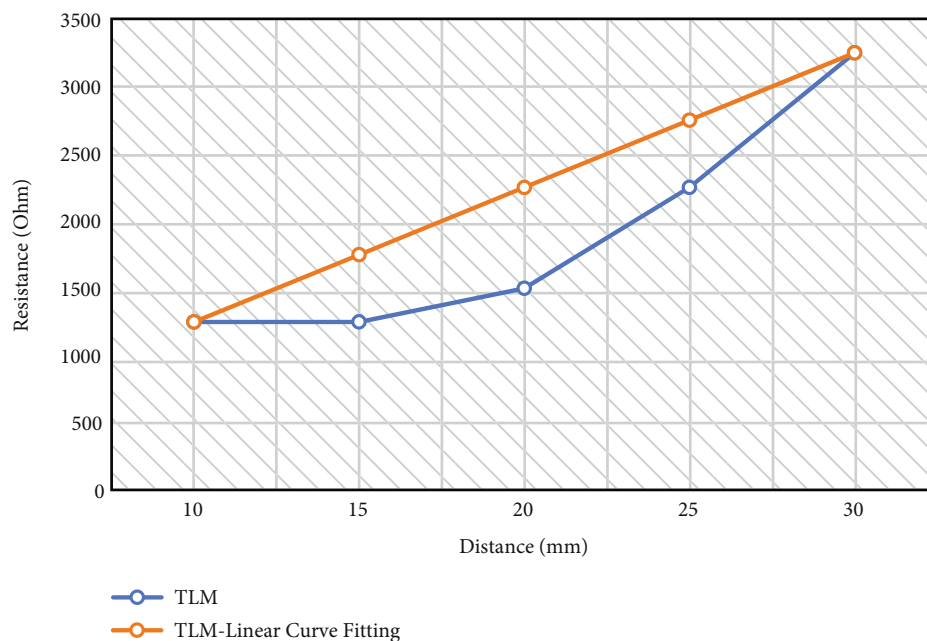


FIGURE 10: Measurement results with TLM-linear curve fitting.

of that of microfabrication. To enhance the resolution and flexibility of composite-based interconnect fabrication, the screen-printing process is being improved. The polyimide must undergo etching in O_2 plasma setting as well as to wet etching the pattern metal layer. As the interconnects must be transmitted from such a carrier semiconductor to polymer substrate, the fabrication process becomes much more complicated. The synthesis of composite-based ink for screen printing interconnections is a future goal. Screen printing cannot be performed using the present PEDOT:PSS composite. On the basis of the composite's encouraging electromechanical responses to strain, more advancements in size reduction are envisioned. The integration of stretchy interconnects with multiple sensor modulus in a greater sensing density is another goal of future research. The sensors are now integrated separately, which takes a lot of time and makes the procedure less reproducible. For the integration of various sensors, the more efficient approach needs to be investigated.

Data Availability

Data supporting this research article are available from the corresponding author or first author on reasonable request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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