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Manufacturing Wearable Electronics by Direct Copper Electrodeposition

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ABSTRACT

In the modern world, wearable electronics are potentially intensifying a transformation of the military, aerospace, medical, industrial, and commercial industries. However, manufacturing electronic circuits or devices directly on fabrics is a major challenge. In this study, we have addressed the issue by developing a two-step manufacturing process by coating a piece of polyester fabric with lignin. Initially, a conductive template was achieved on the fabric after laser-burning lignin. In the second step, using an in-house design printer, copper was electroplated over the conductive template to achieve a very low resistive circuit layout. The conductivity measurements were monitored before and after destructive tests such as bending, rolling, washing, and adhesion tests to prove remarkable stability and flexibility. The quality of the conductive patterns was examined through scanning electron microscopy (SEM). Furthermore, the copper deposition method was applied to solder a light-emitting diode (LED) to the applied circuit layout on the fabric. The proposed two-step manufacturing method has a promising potential to fabricate reliable and leading-edge wearable electronics for various applications.

Keywords: Copper electrodeposition, Electroplating, Laser-burned lignin, Polyester fabric, Smart textiles, Wearable electronics.

1. INTRODUCTION

Since mankind first embarked on the path of civilization, technology has progressed immensely. However, the entire world has been revolutionized at an exceptional rate with the recent comprehensive innovations of semiconductor chips and electronic devices such as computers, tablets, and wireless sensors. One of the evolutions from the repeated advancements in science is wearable technology¹. Even though there isn't a clear definition in most literatures, wearable technology can be defined in the simplest form as the technological devices that are worn on a particular part of the body of a user². Devices like wristwatches were considered wearable technology, and they have existed for centuries. Only lately have they become favored, popular, more functional, and fashionable in different forms like smart watches, calculators, and many more³. Now that technology has taken a turn to include more electronics, they are available in the form of eyeglasses, watches, wristbands, rings, jewelry, shoes or even clothing⁴.

“Smart Textiles” or “Flexible Wearable Electronics” point out to an extensive specialty of products with extensive functionality and usefulness of fabrics, where these woven or knitted structures can interact certain users or the environment. This technology can perform multiple tasks by adding numerous electronics (like sensors) to learn, sense and react, or solely send and receive information. Smart textiles are being widely used in various industries such as medical to monitor health data of patients, perform non-invasive procedures, or perform examinations such as echocardiogram (ECG), electroencephalography (EEG), and electromyography (EMG). They are also used in the military and aerospace industries permitting soldiers and astronauts to support multiple devices in their vests to complete their responsibilities⁵.

The importance of wearable electronics is crucial as it can be used to monitor the health of an elderly or someone with diseases, alerting doctors/caregivers about their patients' overall health, mental health, assistance in emergency management with the help of the sensors and or devices embedded in their smart textiles and self-connecting them with a software that facilitates communication, data-exchange, and access to information in real-time⁶. Smart textiles have been developed in different forms such as sewing, weaving, embroidery, knitting, spinning, coating, printing, chemical treatment, and many others. Many techniques of embedding electronics into fabrics include conductive fibers, treated conductive fibers, conductive inks, inkjet printing, laser burning. While all these methods exist, they

have their pros and cons, some methods can be conductive but not flexible, while some would not be cost-efficient or have higher resolution⁵. Some of the techniques have been discussed below.

Sewing electrically conductive polymers consisting of micro and nano particles in the form of threads or using conductive wires or threads such as copper wires or polyvinyl alcohol (PVA) threads can prove to be a very basic and low-cost technique of obtaining wearable electronics. While there are many patterns that can be sewn on different fabrics, being able to obtain flexibility can be very challenging. Sewing proves to have good resolution and distribution of electronics, however, being able to solder surface mount devices (SMDs) could be one other limitation that can be faced in this technique^{7, 8}.

Different techniques such as woven, sewing, dip coating, spraying, plating, chemical vapor deposition (CVD) with conductive fibers such as carbon nanotubes (CNTs) as a single-wall (SWCNT) or multi-wall (MWCNT) film^{9, 10}, Zinc Oxide (ZnO) nanowires^{11, 12}, etc. have widely been popular as they have very good flexibility and high resolution. Eventually, they are not as cost-effective, and cannot handle soldering SMDs as well.

One of the popular printing processes applied by researchers is inkjet printing, where chemically prepared solution-processable inks made of micro and nanomaterials are directly deposited onto certain locations of the substrate, proving potential to fabricate flexible textiles. Conducting the inkjet printing can lead to advantages such as having programmable, color changeable, higher resolution patterning, fast throughput, no-contact printing to minimize contamination, and cost-effective technique for small sized fabrics. However, there are limitations to this process, where only highly soluble, low temperature inks can be used. Using low boiling point solvents is required to anneal for processing in solvent removal when printing textiles or on plastics. The cost could increase for mass production. Obtaining uniform and good conductivity, higher resolution and being able to solder SMDs are also a few of the limitations^{13, 14}.

As a solution to these challenges, we suggest using laser-burned lignin on fabric and electrodepositing copper via a technique known as Hydrogen Evolution Assisted (HEA) electrodeposition to overcome the limitations of low conductivity and form cost-effective uniform and longer interconnects. One of the reasons we use lignin in this study is that lignin is one of the most redundant, yet abundantly found polysaccharides, produced by plants, providing a high quantity of carbon. Plants produce the lignin by photosynthetically fixing carbon dioxide to reduced sugars, that convert to syringyl (S), guaiacyl (G) and monolignols hydroxyphenyl (H). The process of extracting the lignin into cellulose fibers is known as pulping. This chemical pulping provides us with kraft lignin with low sulfur content¹⁵. We use kraft lignin mixed with deionized water (DI water) as an ink coated over polyester fabric samples, laser burning them with a carbon dioxide type laser engraving equipment and electroplating them to grow copper. The copper electroplating or electrodeposition process is a kind of manufacturing process used to ensure the passing of current throughout the pattern of deposition. Electrodeposition is widely used to fabricate printed circuit boards (PCBs) and many other electronic elements¹⁶. This method proves to be cost-effective and produces conductive patterns that can be used as longer interconnections to connect multiple sensors or electronics for efficient smart textiles with low resistance.

2. EXPERIMENTAL SECTION

2.1 Materials

Kraft Lignin in the form of brown to black powder with low sulfonate content was purchased from Sigma Aldrich (CAS No: 8068-05-1) and used for the lignin solution. A white 100% polyester upholstery fabric was purchased from Kovi Fabrics (Pattern No: E3492) to be used for all research conducted.

Copper Sulfate (CuSO_4) (CAS No: 7758-98-7) and Sulfuric acid (H_2SO_4) (CAS No: 7664-93-9) were both purchased from Sigma Aldrich to prepare the electrolyte solution.

2.2 Preparation of the Lignin Solution and HEA electrolyte

Two grams of lignin was added to 10 mL deionized water (DI water) and stirred for about five minutes till all the lignin dissolved. We found the best results by depositing 260 μL of the solution on the polyester fabric of 45 mm length x 20 mm width dimension by pipetting it and left to dry for 24 hours (Figure 1a). The HEA electrolyte was an aqueous solution prepared with 0.47 M CuSO_4 and 1.5 M H_2SO_4 and mixed with 10 mL of DI water for 5 minutes until thoroughly dissolved¹⁷.

2.3 Lasering Lignin and Electrodepositing Copper

The lignin coated fabric after being dried was lasered in a four-probe pattern with four pads A, B, C and D to examine the conductivity. These pads, each arranged every 10 mm and sized to measure 8 mm x 4 mm were designed on the LaserDRW software (pattern in Figure 1b). The lasering was conducted by placing the fabric in the OMTech 40W CO₂ Laser Engraving Machine (Model No: SH-G3020), setting the laser power at 5% and pattern was rasterized twice. Once the lignin was carbonized by the lasering process, we placed the sample in an in-house designed electroplating printer for metalizing the surface of the conductive laser pattern. The copper electroplating printer design consists of a syringe pump necessary to feed the electrolyte at a controlled rate to the printer nozzle via a transparent tube. This printer nozzle was a 2 mm diameter copper tube contributing as the anode. The nozzle was mounted on the Genmitsu CNC 3018-PRO router kit GRBL control 3-axis machine, XYZ working area 300x180x45 mm stage. The laser-burned conductive template on the fabric was placed on the printer stage. The starting point was considered to be one end of the laser-burned track (pad D) and was connected to the potentiostat as the cathode via a piece of copper tape. We positioned the nozzle to be close to the copper tape with about 2-3 mm space from the surface of the fabric. A 5 mL syringe is filled with electrolyte and placed on the syringe pump. The infusion rate of the pump is set at the rate of 8 mL hr⁻¹ to form a droplet standing between the copper tube nozzle acting as anode and the conductive pattern on the fabric acting as cathode for the localized electroplating. The cathode was applied to a fixed voltage of 3V with respect to the anode. Once the copper growth was observed on the fabric, we moved the nozzle over the fabric to follow the conductive pattern to the end point (pad A). The nozzle movements were controlled manually through the electronic console of the XYZ stage. As we also see in Figure 1c, the printer was furnished with a microscope camera allowing us to monitor the quality of the copper growth and adjust the speed of the printing. Once the copper was deposited on the lignin burned fabric, we allow the fabric to dry for 24 hours to set them for various characterizations.

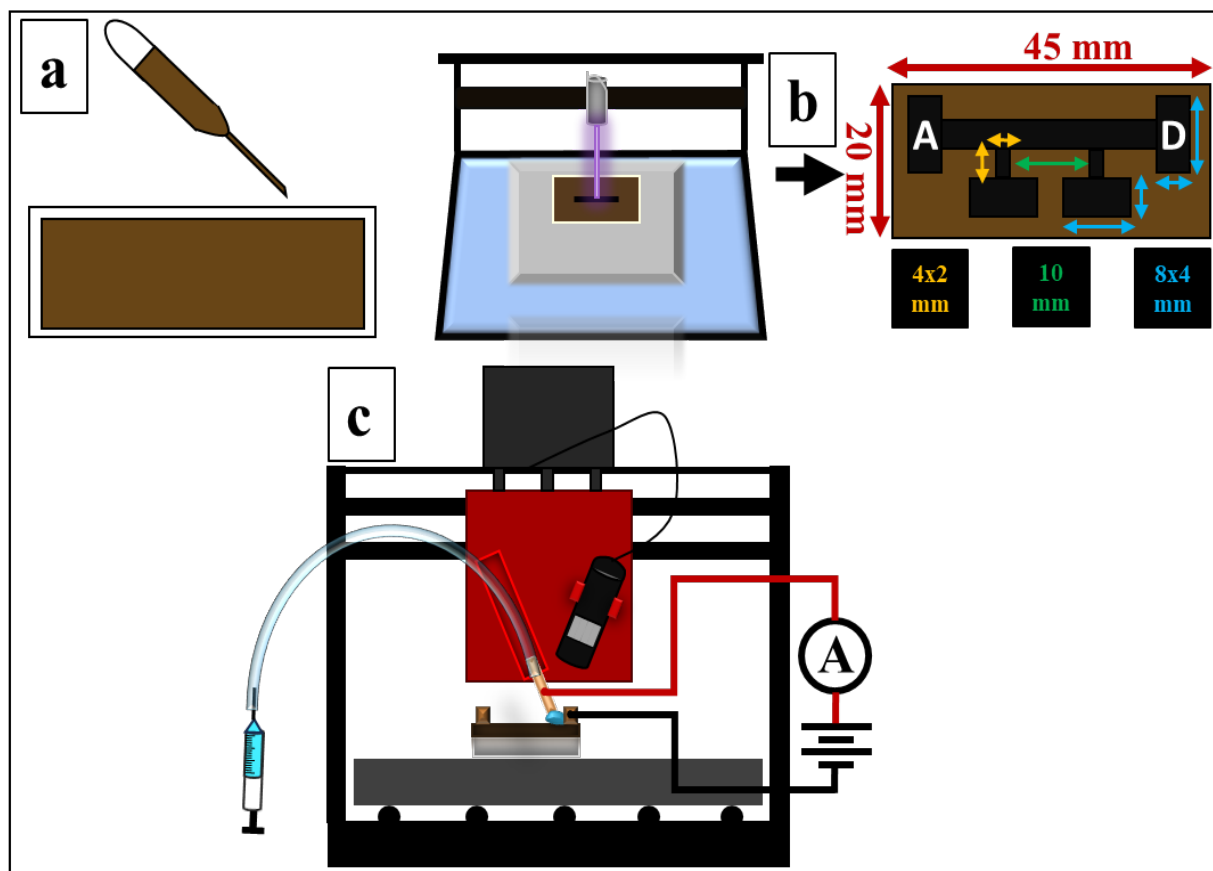


Figure 1. Schematic diagram (a). The process of coating lignin on polyester fabric (b). Laser engraved 4-point probe pattern with given dimensions (c). Electrodepositing copper on the laser-burned lignin with copper tapes.

2.4 Characterizations of the Laser-burned Lignin and Electrodeposited Copper

A four-point probe method using Keithley 2602A system SourceMeter instrument was used to measure the conductivity of the laser-burned lignin and the electrodeposited copper. The average conductivity reading data of each sample was recorded by repeating the process several times. The Hitachi SU-70 high resolution field emission scanning electron microscope (SEM) was used to perceive the structures of the samples. All the experimental, stability and flexibility results are discussed in the following section below.

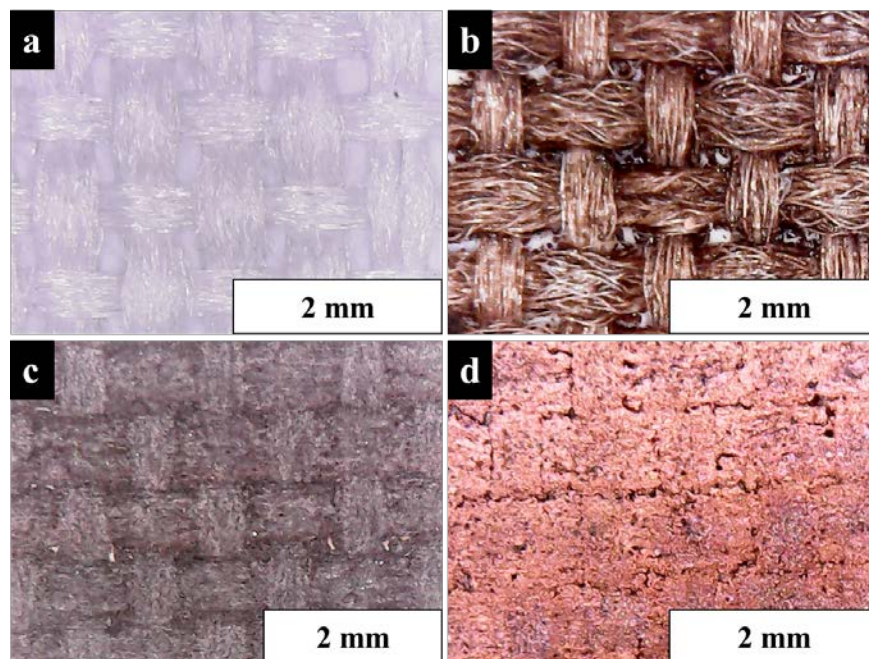


Figure 2. Optical microscopic images (a) bare polyester fabric (b) fabric with coated lignin (c) lignin on the fabric after being lasered (d) electrodeposited copper on the conductive laser pattern.

3. RESULTS AND DISCUSSIONS

3.1 Manufacturing results

The surface morphology and structures of the polyester fabric, lignin coated on the fabric, laser-burned lignin, and copper electrodeposited on the laser-burned lignin were all observed using a USB pluggable digital microscope with a magnification of 20X to 800X. Figure 2 exhibits the optical microscopic images of each step in the process performed. Figure 2a signifies the microscopic image of the bare polyester fabric purchased from Kovi Fabrics. Once the 260 μ L lignin solution is coated on the fabric, the microscopic image Figure 2b represents the dried fabric with the coated lignin. Figure 2c reports the optical microscopic image of the lignin on the fabric after being lasered and patterned with the conductive path. The copper growth on the conductive laser-burned pattern by HEA electrodeposition is represented by the optical image in Figure 2d once the copper grows and the sample is dried.

Figure 3 below exhibits the potentiostat data of the current in mA vs. time in s when the copper was being electrodeposited over the four-probe laser-burnt pattern between pads A and D. The recorded current in Figure 3 shows that during the first 6 seconds, the current remained 0, meaning that the electrolyte droplet hadn't made contact with the fabric. At the 7th second, the current dropped to about 143 mA, indicating that the electrolyte drop was in touch with the fabric, and the process of HEA electrodeposition had begun. In a span of about 2-3 seconds, as soon as the copper started to grow on the laser-burned lignin, the current spiked to about 47 mA. As the optical image in Figure 2d shows a relatively uniform coating of copper deposited over the pattern, the nozzle was slowly moved over the sample as the copper grew. The speed of the nozzle was manually controlled to maintain the electrochemical current in the range of 32 mA to 50 mA. The inset picture in Figure 3 shows the copper tube as the printer nozzle that was the anode, and the electrolyte droplet on the fabric with the cathode being the copper tape on the lasered

conductive path. After successfully electroplating, the samples were dried in a desiccator and were then characterized to further investigate the nanostructure of the layers.

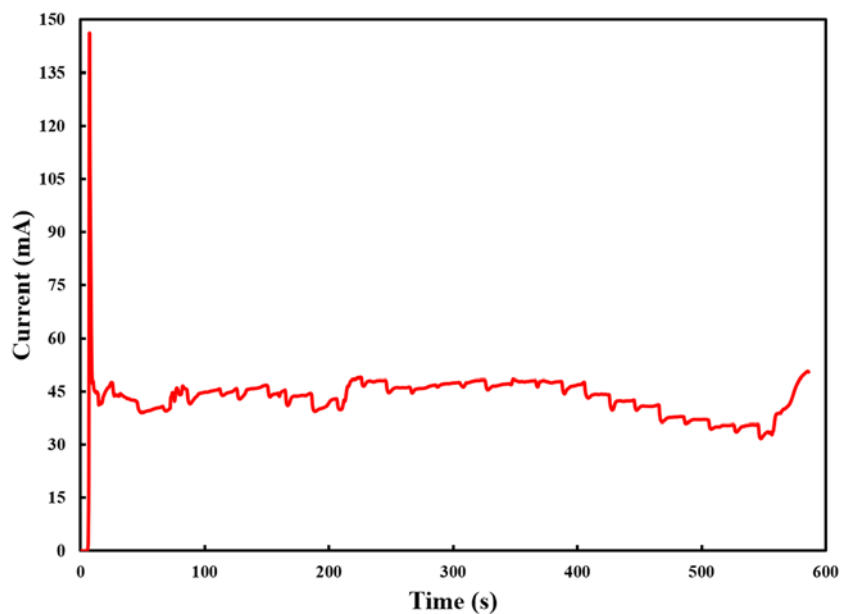


Figure 3. Potentiostat data of electrodeposition current at 3V as the nozzle was moved at a rate of 70 $\mu\text{m}/\text{sec}$.

3.2 Characterization results

SEM with the Hitachi SU-70 instrument was used to evaluate the surface morphology of the electroplated copper grown on polyester fabric as seen in Figure 4, depicting the magnifications at 1 mm, 300 μm , 100 μm and 50 μm .

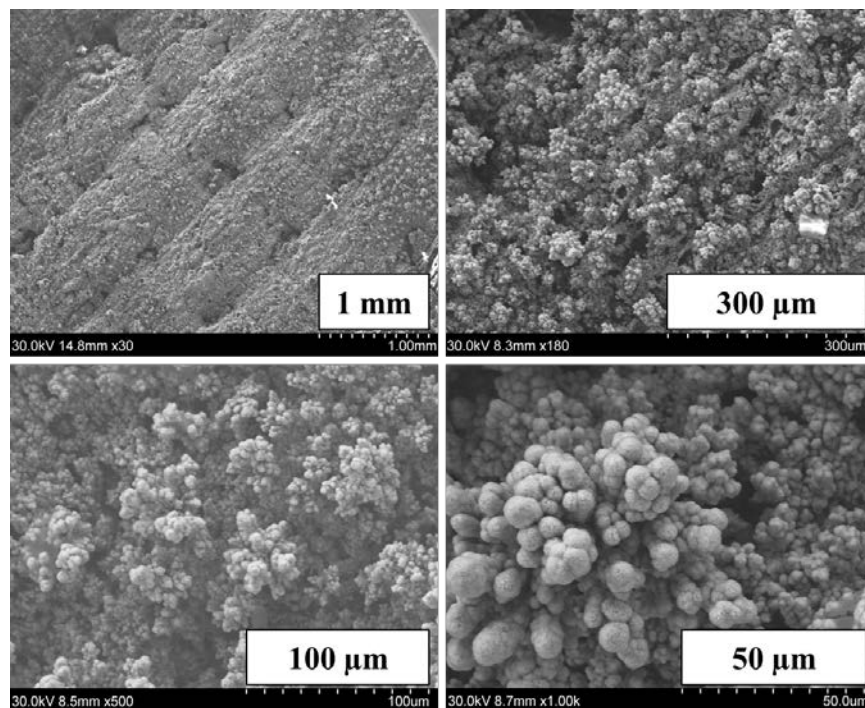


Figure 4. Scanning Electron Microscopic (SEM) images of electrodeposited copper grown over lasered lignin on polyester fabric.

3.3 Stability and Flexibility testing results

When the samples were tested using the four-point probe using the Keithley 2602A instrument, the measured resistance value was found to be $196\ \Omega$ for the laser burned lignin sample. Once electrodeposited with copper, the resistance value was $11.32\ \Omega$. After measuring the resistance, the stability tests were performed.

We tested the mechanical stability and flexibility of the pattern by conducting bending and rolling tests. We used a 3D printed cylinder of 1.27 cm diameter to roll on the samples back and forth 10 times to test for any change in resistance values, resulting in no change. We also utilized two different 3D printed semicircle configurations of 2.54 cm and 3.81 cm diameter to bend the samples on and simultaneously recorded the change in resistance values resulting in no difference.

Once these tests were repeatedly performed, we washed the samples by sinking them for five minutes in a beaker filled with 200 mL of DI water and drying them. We remeasured the resistance value after the samples were dried only to find the resistance value to be increased by $\sim 0.71\ \Omega$ for the electrodeposited copper sample.

Following the stability tests, we tried destructive tests, such as adhesion test to observe the adhesive nature of the samples. To perform this trial, we applied the Scotch 3M brand Satin Finish Giftwrap Tape over each sample and added a weight of 1.4 kg on the samples for two minutes. We then ripped the tapes in two different motions – swift and slow, to compare how much lignin or copper would come off the fabric and how that would affect the resistance values. Ripping the tapes in either swift or slow motion did not take off much of the laser burned lignin or copper growth and did not affect the resistance of the samples.

3.4 Application – Integration of a light emitting diode (LED)

To prove our hypothesis of achieving longer interconnects while having low resistance, we performed an experiment as a part of our study. We lasered a simple conductive path of two conductive strips of 4 mm x 15 mm with a gap of 1 mm between them. We used a dot of hot glue and attached an LED in the middle of the laser-burned fabric right before electrodepositing copper. Once it dried, we carried out our process to grow copper on the lasered pattern till it reached LED terminals from both sides, enough to create junctions from both ends. We allowed the sample to dry in the desiccator for 24 hours. We then connected the sample to an external Arduino board and the LED lit throughout the path where copper was grown as represented in the form of optical microscopic images in Figure 5.

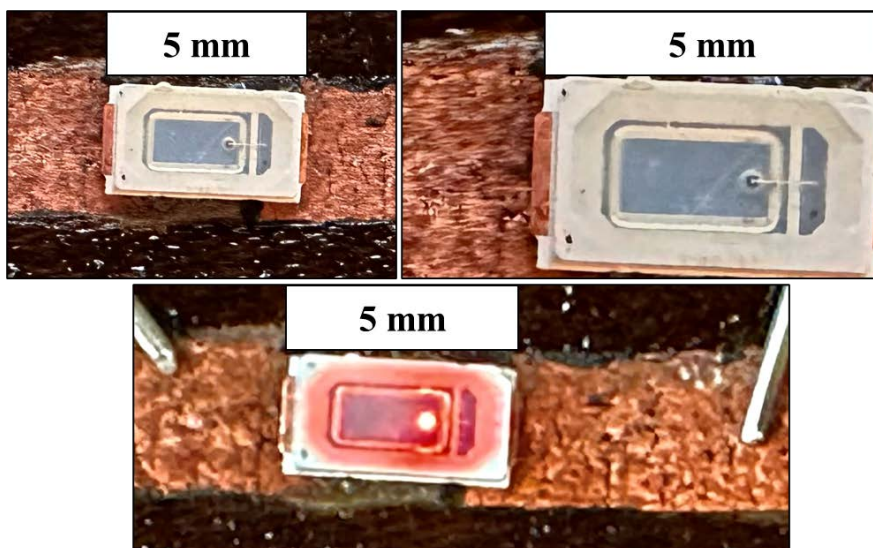


Figure 5. Optical microscopic images of copper electrodeposition (top left), LED junction created by copper growth (top right) and LED lighting up by the power of an Arduino board (bottom).

The sequential process of laser burning lignin and electrodepositing copper on fabric directly shows promising results in terms of low resistivity, flexibility, and stability. This cost-effective process can be used to solder sensors and attach other electronic gadgets to manufacture wearable electronics with upgraded functions.

4. CONCLUSIONS

To summarize, accomplishing the laser-burning process to fabric with the help of lignin, and growing copper on the lasered path can be rewarding and beneficial for numerous applications, to attain longer interconnects with lower resistance and installing multiple devices/sensors whilst low resistance. Obtaining a high-quality graphene/graphite can be easily achieved by laser burning the lignin solution on the fabric directly, thus proving to be a favorable solution to issues occurring with respect to stability and flexibility. To achieve a uniform copper growth rapidly when compared to conventional electroplating, the HEA electrodeposition process shows favorable outcomes, yielding state-of-the-art futuristic wearable textiles.

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