Screen Printing Silver Stretchable Conductive Paste to High Density Synthetic Fabric

A Senior Project
Presented to

The Faculty of the Materials Engineering Department
California Polytechnic State University, San Luis Obispo

In Partial Fulfillment
Of the Requirements of the Degree of
Bachelor of Science, Materials Engineering

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DuPont & NextFlex

Abstract

This project investigated the viability of screen printing stretchable silver conductive paste directly onto fabric and how the resistance changed under cyclic mechanical loading. The paste tested was DuPont™ PE873 stretchable silver conductive paste, which forms a stretchable conductive path by suspending silver flakes in elastomer that can be elastically strained along with the underlying substrate. The silver pastes were printed directly onto two different high-density synthetic fabrics of different weaves. Other samples were prepared by first printing a base layer between the silver paste and the fabric. One base layer was a solvent-based dielectric (DuPont™ ME776) and a stretchable carbon conductor (DuPont™ PE671). This project determined that printing onto either base layer had a significantly smaller change in resistance under cyclic tensile tests than printing onto the fabric directly. Bend tests also revealed the possibility that the rate of change of strain had a higher impact on the change in resistance than the strain itself.

Keywords:

Screen printing, Stretchable conductive paste, Flexible electronics, Silver flakes, Direct, Fabric, PET, Nylon, Simple weave, Satin weave, Dielectric, Carbon paste, Resistance, Tensile test, Bend test, Reliability, Strain

Acknowledgements
We would like to thank our contacts industry contacts: Dr. Homer Antoniaidis, Jeffrey Dee, and Dr. Daniel Inns from DuPont and Dr. Kollengode Narayanan, Mike Mastropietro, and Dr. Jason Marsh from NextFlex. We appreciate this opportunity and your guidance and patience throughout this project. We would also like to acknowledge Prof. Jianbiao Pan and Josh Ledgerwood from Cal Poly's Industrial Engineering department and Prof. Malcom Keif and Prof. Xiaoying Rong for your expertise, training, and advice, as well as allowing us to use your facilities. Next, we would like to thank the Materials Engineering Department: Prof. Victor Granados for taking your time to help us with the SEM, Lisa Rutherford for purchasing items and being patient with our constant pestering, and all the other professors for answering any questions we had. Finally, we would like to thank Prof. Linda Vanasupa, our senior project advisor, for helping us work through any uncertainty or problems we faced and supporting us throughout the year.

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1. Problem Statement

Currently, wearable electronics technology is largely limited to making flexible designs around rigid electronics. To print a flexible circuit into fabric requires that the conductive paste is printed onto a plastic like thermoplastic polyurethane (TPU) and then laminated onto the fabric. This project investigated the viability of directly printing a silver, stretchable conductive paste to commercially-available, synthetic, fibrous fabrics. Change of resistance was measured during repeated tensile loading to 10% strain and a modified crease test with a minimum bend radius of 1mm. Scanning electron microscopy (SEM) was used to characterize the differences in silver paste after mechanical testing.

2. Background

The wearable electronics market is moving towards integrating printed circuits into fabric. Currently, the circuit needs to be printed onto a plastic substrate, often TPU, then laminated on the fabric. One way to reduce production time and costs is to directly print the circuit onto fabric. The integration of printed circuits into smart textiles requires the ink to withstand repeated tensile strain during use. This includes repeated putting on and taking off of the garment as well as stretching during use (e.g. running, weight lifting). Studies have been done on the effects of repeated tensile loading of stretchable and non-stretchable conductive inks on stretchable substrates.

2.1 Tensile Testing of Conductive Ink Printed on Fabric

Most tensile research on non-stretchable conductive inks focuses on the change of resistivity during tensile loading. This is because most stretchable inks will fail after multiple loadings due to cracking or not adhering to the substrate. A study done in 2010 at Tampere University of Technology in Finland looked at the effects of strain on the conductivity of a polymer thick film (PTF) ink on three different substrates. The ink was a polymer matrix with conductive silver flakes. The substrates stretched; the ink did not. The three substrates used in this study were: stretchable polyvinylchloride (PVC), commercial elastic polyester/spandex fabric, and commercial elastic fiber without spandex. The ink was screen printed onto each substrate then strained to from 0% to 50%. The samples were tested at 0%, 5%, 10% and then every 10% strain thereafter. The PVC samples were stretched three times. Two-point resistance testing was used during straining and during recovery. A 50 mm/min strain rate was used. The study concluded that the PTF ink maintained conductivity under strain. The resistivity of the ink correlated with the applied strain.[1] Scanning electron microscopy (SEM) was used to determine the mechanism of change between the PVC and fabric substrates. It was concluded the behavior of the ink on PVC acted differently than the fabric substrates, which played an important role in the changes of resistivity of the ink.[1]

2.2 Stretchable Conductive Ink in Tensile Loading

Stretchable conductive inks are a relatively new development. In a study published in 2016 in the Applied Physics Letters, researchers at Flex and the Center for Advanced Life Cycle Engineering of the University of Maryland conducted tensile tests on two generations of

stretchable conductive inks. The first-generation ink (Gen 1) was 178X, which was chosen as the baseline. The later-generation ink (Gen 2) was N 6301X. Both inks were screen printed on a TPU substrate and low-temperature sintered. The samples were strained at a rate of 0.1 mm/s. Resistance was measured during straining and after cyclical loading. The Gen 1 sample was strained until 57%. It showed an increase in resistance from 1.9 Ω to 1400 Ω.[2] Most of the increase in strain occurred at ~40% strain.[2] The Gen 2 ink was strained to 100%, which correlated with an increase of resistance from 4.8 Ω to 835 Ω.[2]

Gen 2 ink under cyclical loading was significantly better than the Gen 1 ink. The Gen 2 ink remained stable up to 500 cycles of 20% strain loading; the resistance increased from 2.6 Ω to 50.9 Ω. After 750 cycles, the resistance increased unpredictably.[2] Comparatively, the Gen 1 ink retained conductivity for only 30 cycles of 20% strain.5 The study concluded that though the Gen 2 ink was superior in both tests, more development is needed.[2] Most stretchable electronics require at least 2000 stretch cycles at 20% strain, which neither ink could fulfill.[2]

3. Methods and Procedures

3.1 Materials

There were six permutations of samples made for this experiment. Two fabric substrates were used. On the fabric, the stretchable conductive paste was printed directly on the fabric, on a dielectric base layer, or on a conductive carbon paste base layer.

3.1.1 Fabrics

Two fabric substrates were used. The criteria for the fabrics were that they were high density, synthetic, and fibrous fabrics. One substrate was a five-harness satin weave composed of 100% PET fibers (Figure 1a). The second substrate was a simple weave that was composed of 89% PET and 11% nylon (Figure 1b).

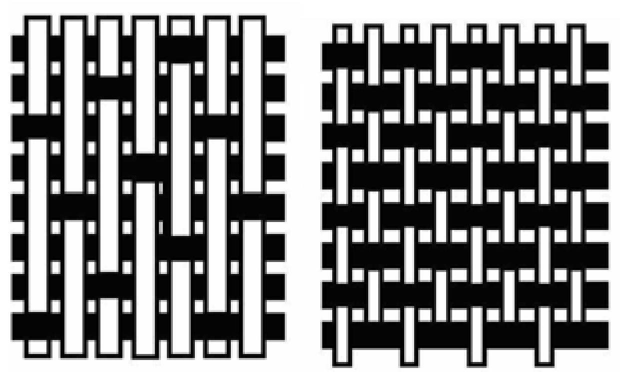


Figure 1*: (a) Diagram of 5-harness satin weave used in the 100% PET substrate. (b) Diagram of simple weave used in the 89% PET 11% Nylon substrate.*[3]

Scanning electron microscopy was used to image the fabrics and estimate the apparent "gap size." The gap size was defined as the space between overlapping roving of fiber. The satin weave had no visible gap, while the simple weave had a 100 µm (Figure 2).

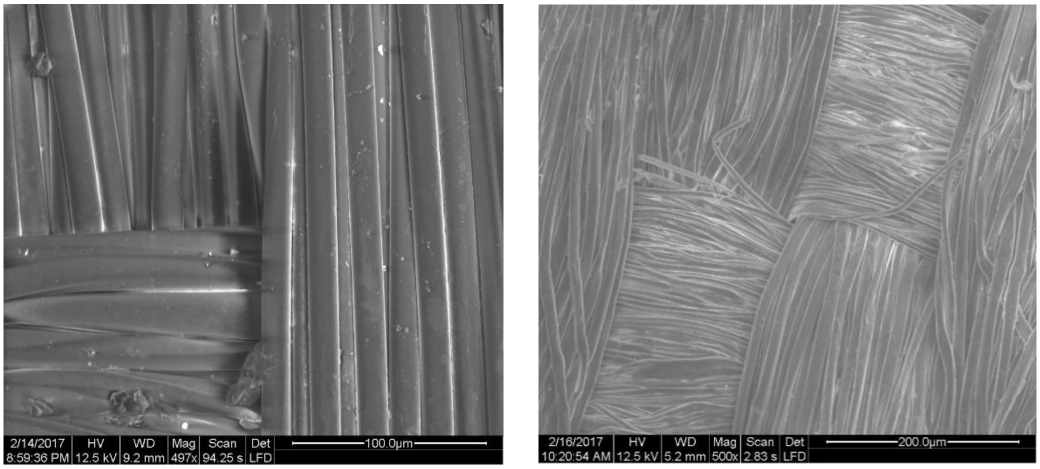


***Figure 2**: Scanning electron microscope images of (left) satin weave substrate (right) simple weave substrate to show the apparent gap size of the fabrics.*

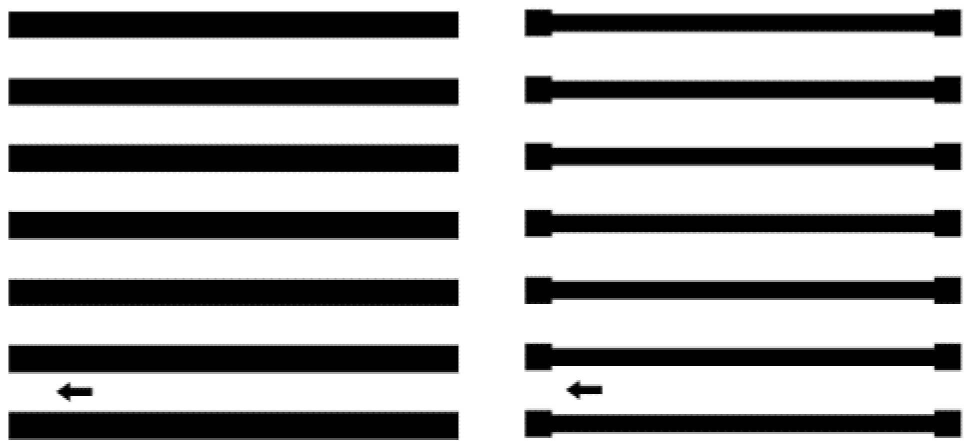
3.1.2. Base Layers and Stretchable Silver Conductive Paste

The base layers and silver conductive paste were provided by DuPont. The two base layers used were a carbon conductive paste (PE671) and a solvent-based dielectric (ME776). The silver conductive paste was a third-generation stretchable conductive paste (PE873). After curing, the solvent is evaporated leaving silver flakes as the conductive path, which is suspended in a stretchable resin that provides adhesion to the substrate and cohesion of the silver flakes. The silver-flakes act like platelets that slide over each other when stretched. This allows the conductive path to be maintained while being stretched.

3.1.3. Sample Fabrication

The samples were fabricated using a Dek Horizon i03 screen printer. A vacuum table was used to stabilize the fabric while printing. Each layer was manually aligned on the vacuum table using the upper left corner of the sample. The front and back squeegee pressures were 2.6 kg and 4.6 kg respectively. The print speeds used were 50 mm/s forward, and 25 mm/s backward. The separation speed was 1.0 mm/s with a separation distance of 3.0mm. Each layer was cured at 130°C for 10 minutes in an Espec box oven.

The traces were printed using custom stencils made by Sefar. The screens were a 0.0014" wire diameter with a 200 wires/inch mesh count. The emulsion used was Sefar's E80 with a 20 µm thickness. Two traces were designed; one with 6mm bars for the base layer (Figure 3a), and one with 4mm bars with 6mm connecting points for the silver paste (Figure 3b). The traces were cut into strips with one bar per sample for testing.



(a) (b)

Figure 3*: a) Base layer trace composed of seven 102mm x 6mm lines b) silver paste trace composed of seven 102mm x 4mm lines with 6mm x 6mm connecting points.*

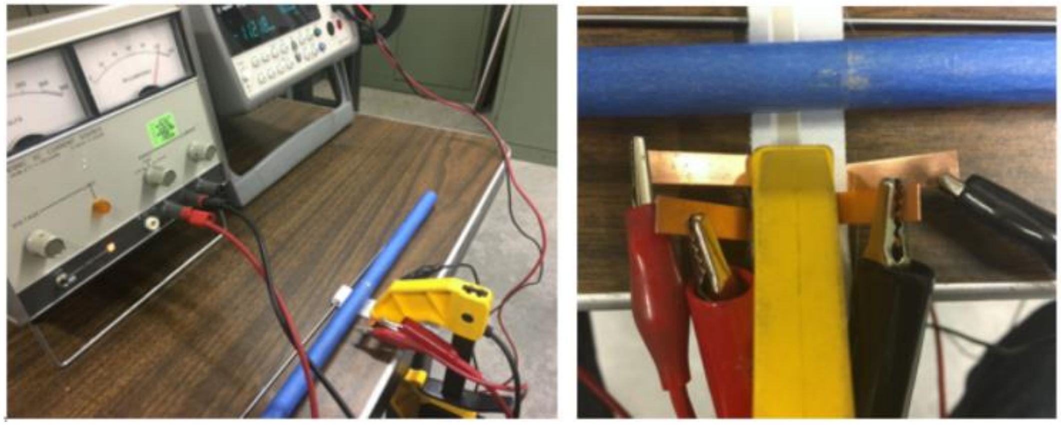
3.2 Tensile Test Procedure

Tensile tests were performed using a Mini Instron55 tensile tester. The samples were strained to 10% elongation at 100 mm/min for 50 cycles. A macro was used to automatically return and start the Mini Instron 55 tests in the bluhill 3 software. This allowed the tests to be repeated once every 10 seconds. An Aligent 34405A multimeter and HP 6186C DC current source were used to measure the change in resistance over the repeated strains. The resistance was measured using the four-point probe method by inducing a 10mA current while measuring the change of voltage using the voltmeter. The resistance was then calculated using the current and voltage. A Matlab code was used to data log the resistance using the USB connection in the multimeter (Appendix A).

3.3 Bend Test Procedure

Before setting up the bend test samples for testing, the resistance between the terminals was recorded by using a digital multimeter. The bend tests were then setup by folding a sample in half so that the print was on the outside of the fold and clamping the end to a table. Between the clamp and the sample were two copper strips that were connected to both terminals to form a four-point probe test like the one in the tensile test (Figure 4). The resistance was recorded and compared to the resistance measured by the digital multimeter before setting up the test.

A 2mm diameter steel rod was inserted into the fold. Then, a 13mm diameter rod was used to apply pressure to the sample by rolling it towards the 2mm diameter bend. When the roller could no longer roll freely, the sample would be bent at a radius of 1mm. The pressure was relieved when the roller rolled back, and the sample would return to a natural bend radius greater than 1mm. This was repeated in intervals, with five seconds of pressure and five seconds without pressure for twenty-five cycles for each sample. Like in the tensile test, resistance was measured for at least a minute after the cycles were completed.



(a) (b)

Figure 4: a) Base layer trace composed of seven 102mm x 6mm lines b) silver paste trace composed of seven 102mm x 4mm lines with 6mm x 6mm connecting points.

4. Results

4.2 Tensile Test Results

The maximum resistance achieved and minimum resistance from each run were subtracted (Table I). The change in resistance was used because accounts for the effects resistance has on design constraints of a circuit. The simple weave with only silver paste showed the largest increase in resistance. The maximum change in resistance achieved was 2459 Ω, with an average of 1575 Ω. The second highest maximum change in resistance was the satin weave with only silver paste as well. The satin weave samples with dielectric and carbon had the lowest change in resistance.

Table 1: Difference in Resistance Values from Cyclic Tensile Tests

Samples	Difference in Resistance (Ω)						
	1	2	3	4	5	Average	Standard Deviation
Simple/Dielectric	78.07	83.28	69.79	64.61	42.04	67.56	15.99
Simple/Carbon	28.20	63.95	62.85	67.88	62.13	57.00	16.25
Simple/Silver-Only	2459	1166	2081	1097	1074	1575	648.9
Satin/Dielectric	21.36	20.05	20.59	21.24	22.05	21.06	0.7654
Satin/Carbon	16.56	23.89	22.47	24.70	20.21	21.57	3.276
Satin/Silver-Only	154.9	133.3	97.65	81.75	84.58	110.4	32.23

4.3 Bend Test Results

For any given bend test, the resistance values before and after the test were setup varied significantly. Since a consistent resistance value could not be achieved before and after setup, the team hypothesized that slight changes in pressure from the clamp were altering the resistance value of the sample. Changes in resistance values when more pressure was applied in the clamp supported this hypothesis. However, due to the time constraints of the project, the procedure was not altered to regulate the change in resistance. Instead, the procedure was carried out three times for each permutation to observe the nature of how the resistance would change as cyclic bend stresses were applied.

The bend test showed two distinct peaks of resistance between each cycle (Figure 5). These corresponded with when pressure from the roller was applied and relieved. For some cycles, the higher peak in resistance was when pressure was applied; other cycles had a higher peak when pressure was relieved. This behavior was observed in all the permutations of the test.

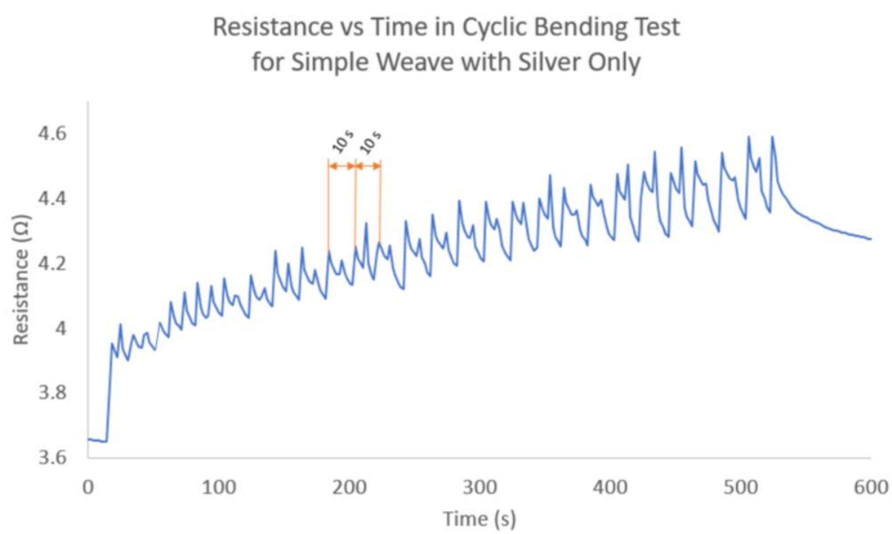


Figure 5: *Resistance vs. time in the cyclic bending test for a simple weave with silver paste only. The twin peak behavior with varying heights was observed in all the samples.*

5. Discussion

5.1 Cyclic Tensile Tests

The increase in resistance over repeated loads can be explained with loss of silver. The equation for resistivity can be rearranged to solve for resistance (Equation 1).

$$R = \frac{\rho L}{A} \tag{1}$$

Where R is resistance, ρ is resistivity, A is conductive path cross-sectional area, and L is conductive path length. With loss of silver, the cross-sectional area of the conductive path decreases, which increases the resistance.

Since the paste only samples were outside the standard deviations of the other samples, those samples were excluded from any statistical analysis. The paste only samples act differently from those with a base layer. Scanning electron microscopy was used to confirm the loss of silver. The paste only samples on both substrates had greater loss of silver than those with base layers (Figure 6). The carbon and dielectric form better substrates for the silver paste to adhere to the surface. The base layers are able wet the fabric better than the silver paste. This flattens the topography and causes superior cohesion between the base layer and the silver paste compared to that of the fibers in the fabric.

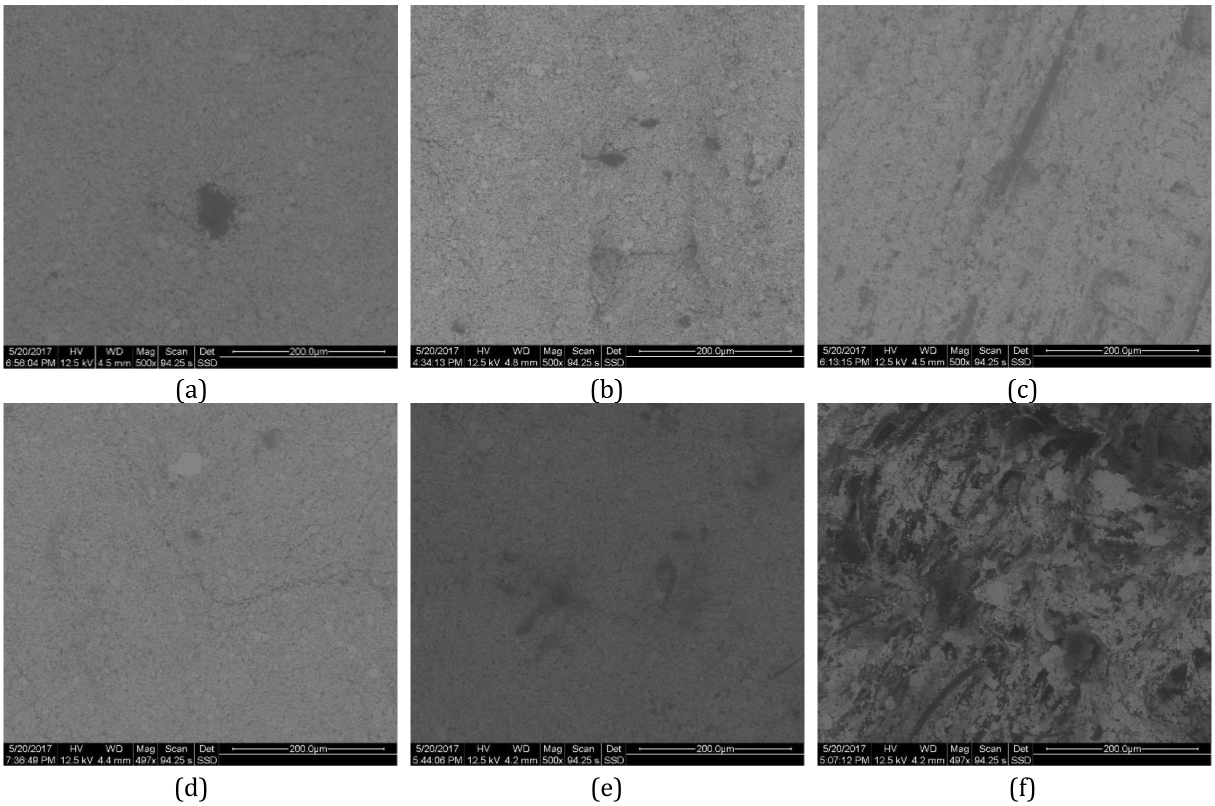


***Figure 6**: Backscatter SEM images at 500x taken in low vacuum mode of a) silver/carbon/satin b) silver/dielectric/satin c) silver/satin d) silver/carbon/simple e) silver/dielectric/simple f) silver/simple after 50 cycles of 10% strain.*

A one-way analysis of variance was performed to compare the effect of cyclic tensile straining on the change of resistance for the carbon and dielectric samples. Which base layer was used did not have a significant effect on the change of resistance to a confidence interval of 95% [F(1,18)=0.2505, p=0.6228]. The substrate had a significant effect on the change of resistance with a confidence of 95% [F(1,18)=59.7538, p=<0.0001]. The effect of the substrate can be explained by observing the strain concentrations of the fabrics caused by the roving overlapping. The areas where the roving overlap cause a strain field as the transverse and longitudinal roving strain each other due to the applied tensile stress. These strain fields cause an increase in resistance. The satin weave has a roving overlap every four roving along the longitudinal shafts, while the simple weave has more overlaps. Thus, the simple weave has more strain fields that interfere with each other, increasing the resistance. This theory also applies to the effects of the base layer. In regions where the

roving overlap, the carbon or dielectric layer relieve a bulk of the strain from the silver paste, reducing the effects of the strain on the resistance.

5.2 Cyclic Bend Tests

The team expected that the resistance of the sample would peak as pressure is continually applied and remain constant while constant pressure is applied. However, peaks of resistance when the amount of pressure by the roller is changed suggests that rate of change in strain had a more significant impact on the peak resistance of the sample. Even if the pressure was being relieved, the change in strain caused the resistance to spike. This phenomenon can possibly be explained using percolation theory. If an electron is travelling along a path of least resistance, there is some ideal path that is in the process of following. In Figure 7, this path is represented by the shorted path from the negative side A to the positive side B (marked with yellow tiles). In reality, the electron is navigating along connected paths between the silver flakes and around voids. As the strain changes, the flakes rearrange themselves, which causes the path of least resistance to change. This can cause an increase in travel time of the electrons from one end of the material to the other (Figure 7), which is interpreted by the four-point probe test as an increase in resistance.

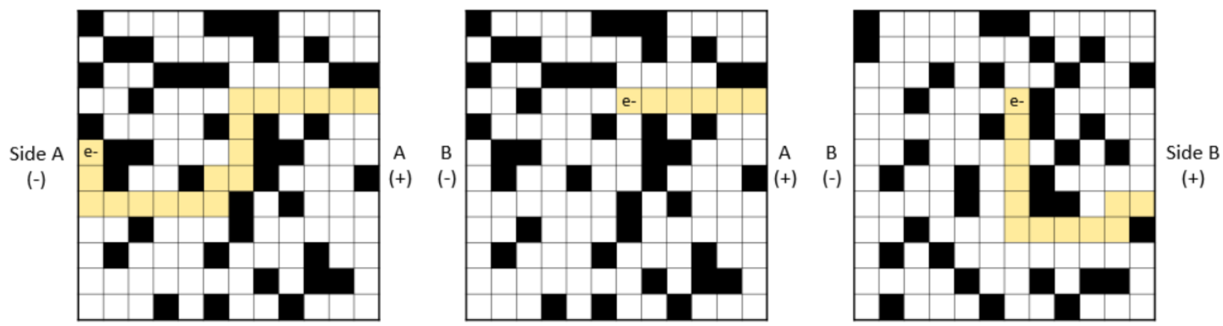


Figure 7: *Percolation theory representation. (a) Shows the shortest path for an electron to travel to the right side along white tiles (representing conductive paths) and avoiding black tiles (representing voids). (b) Shows the remaining path for the electron once it's about halfway to the right side. (c) Shows the new shortest path for the electron after the distribution of voids has been rearranged, forming a new longer path for the electron to travel.*

6. Conclusions

(1) In repeated tensile straining, the silver paste directly printed onto the fabric will have a significantly greater increase in resistance than those printed onto a carbon or dielectric base layer.

(2) In repeated tensile straining, the substrates had a higher influence on the resistance behavior than changing the base layer.

(3) While in bending, the rate of change in strain has more influence in the resistance than the amount of stain applied.

7. Future Work

In the future, it could be valuable to investigate some aspects that this project did not have the time to address. First, without an established bend test procedure for flexible electronics already in place, the procedure used in this study had to be developed by the team. As a result of the clamping method having variable pressure the resistance varied significantly. This fault in the testing procedure could be addressed by using a new jig developed (simultaneously during the course of this project) by the Mechanical and Industrial Engineering departments at California Polytechnic State University, San Luis Obispo to perform bend tests on flexible samples using the same tensile tester used in this study. It would be able to test different bend radii at specific strain rates and apply a constant amount of pressure to the terminals from the leads.

It would also be worthwhile to investigate whether printing all three layers together would have a smaller change in resistance than printing on just one base layer. Since one is a stretchable conductor and the other is a dielectric, it is possible that printing both base layers could improve the performance. A test could also be performed to see if using different screen print settings can produce different or better results under cyclic mechanical loading.

The mechanical testing procedure could also be improved by adding methods to vary the strain rate. This could test the hypothesis that the change in resistance is at least partially a function of strain rate. The tests could also be adjusted to characterize the anelastic behavior of the resistance after cyclic loading (Figure 5).

8. References

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(3) Khatavkar, N.; K., B.; Mcllhagger, R.; Choi, J. H.; Kim, K. S.; Halim, Z.; Hangouet, J. P.; Barbe, N. Composite Materials for Supersonic Aircraft Radomes with Ameliorated Radio Frequency Transmission-a Review. *RSC Adv.* **2016**, *6* (8), 6709–6718.

9. Appendix

Appendix A: MatLab Code for Data Logging

```
clear; %clear variables
clc; %clear window

%% Set-up Multimeter
% Find a VISA-USB object.
obj1 = instrfind('Type', 'visa-usb', 'RsrcName',
'USB0::0x0957::0x0618::TW47230040::0::INSTR', 'Tag', '');
      %VISA-USB address can be found using Agilent VISA or Keysight
Command Expert

% Create the VISA-USB object if it does not exist
% otherwise use the object that was found.
if isempty(obj1)
    obj1 = visa('AGILENT',
'USB0::0x0957::0x0618::TW47230040::0::INSTR');
else
    fclose(obj1);
    obj1 = obj1(1);
end

%% Open object
fopen(obj1);
%% Set the instrument in remote mode
fprintf(obj1,'SYSTEM:REMOTE');

%% Open text document

fileid = fopen('voltagedata.txt', 'w');

%% Set up the figure window
time = 0;
voltage = 0;

figureHandle = figure('NumberTitle','off',...
    'Name','Resistance Characteristics',...
    'Color',[1 1 1],'Visible','off');

% Set axes
axesHandle = axes('Parent',figureHandle,...
    'YGrid','on',...
    'YColor',[0 0 0],...
    'XGrid','on',...
    'XColor',[0 0 0],...
    'Color',[1 1 1]);

hold on;

plotHandle =
plot(axesHandle,time,voltage,'Marker','.','LineWidth',1,'Color',[0 1
0]);
```

```
xlim(axesHandle,[min(time) max(time+0.001)]);

% Create xlabel
xlabel('Time','FontWeight','bold','FontSize',14,'Color',[0 0 0]);

% Create ylabel
ylabel('Resistance in
Ohms','FontWeight','bold','FontSize',14,'Color',[0 0 0]);

% Create title
title('Resistance Characteristics','FontSize',15,'Color',[0 0 0]);

%% Set the time span and interval for data collection
timer = datenum(clock + [0,0,0,0,5,0]);
    % [0,0,0,0,x,0] where x = length of test in minutes
    % x=10 for tensile tests and x=5 for bend tests
timeInterval = 0.000005;

%% Collect data
current = .01;
count = 1;

while datenum(clock) < timer
    time(count) = datenum(clock);
    fprintf(obj1,'MEASURE:VOLTAGE:DC?'); % To measure current the
command is MEASURE:CURRENT:DC?
    voltage(count) = fscanf(obj1,'%f');  %#ok<SAGROW>
    set(plotHandle,'YData',voltage/current,'XData',time);
    set(figureHandle,'Visible','on');
    datetick('x','MM:SS');
    fprintf(fileid, '%6.5f \r\n', voltage(count)/current);

    pause(timeInterval);
    count = count +1;
end

%% Print to document
fclose(fileid);

%% Put the instrument in local mode
fprintf(obj1,'SYSTEM:LOCAL');

%% Clean up the serial object
fclose(obj1);
delete(obj1);
clear obj1;
```