Evaluation of Stretchable Conductive Ink

Weifeng Liu, Ph. D., Jie Lian, Ph. D., William Uy, Zhen Feng, Ph. D.
Anwar Mohammed and Murad Kurwa
Advanced Engineering Group, FLEX International,
Milpitas, CA, USA

Abstract

With increasing popularity and momentum for wearable and printed electronics, stretchable printed ink has become a hot subject for study and application. Circuitry printed from stretchable ink remains conductive even when it is elongated at a high percentage, thus it is useful for many wearable applications where stretching is required.

There have been limited studies on stretchable ink and its stretchability. The characteristics of the stretchable ink and how different factors impact its stretchability are not well studied and therefore unknown. To accommodate the increasing demand for wearable applications, it is necessary to perform evaluations on the stretchable ink.

This paper presents the company studies on stretchable printed ink. A test coupon is designed to prevent instability of connection between test probe and printed ink pad. Different design and test parameters are evaluated to shed light on how they affect the stretchability. The study is very useful to not only optimize the ink stretchability, but also provide insight on how to improve ink chemistry to enable more stretchability.

Key words: Flexibility, Stretchability, Conductive Ink, Printed Electronics, Stretchable Electronics.

Introduction

Stretchable electronics are electrical and electronic circuits and combinations of these that are elastically or inelastically stretchable by a certain percentage while retaining functionality [1]. Stretchable electronics has become a popular subject for study and application, along with wearable electronics, since it attempts to mimic the human skin and body, in being stretchable, whilst retaining full functionality. Stretchable electronics has found many applications for wearables, such as baby sleep monitors [2], stretchable skin medical patches [3], etc.

Three core elements enable the stretchable electronics: stretchable interconnect, stretchable substrate and/or stretchable encapsulant. The stretchable interconnect refers to the conductive trace between two electronics devices. To make the interconnect stretchable, traditionally the conductive trace is designed in a meandering way, such as a serpentine shape, or half sine wave, so that it can be stretched without breaking. However, the meander interconnect takes extra space, this may be challenging for designs with real estate constraints. A new way to make stretchable interconnect is to print stretchable conductive ink on a stretchable substrate. The conductive ink is stretchable even it is printed in straight lines. The stretchable conductive ink can be printed through flatbed screen printing or roll to roll (R2R) screen printing, enabling high volume production. The stretchable conductive ink shows a great potential for stretchable electronics applications. However, as a new technology, few data is available on the stretchability of the stretchable conductive ink. Stretchability test methods need to be developed and validated. Different design and test parameters need to be evaluated to understand how they affect the ink stretchability.

This paper presents the company's first studies on stretchable printed ink. A test coupon is designed to prevent instability of connection between the test probe and printed ink pads. Different design and test parameters are evaluated to shed light on how they affect the stretchability. The study is very useful to not only optimize the ink stretchability, but also provide insight on how to improve ink chemistry to enable more stretchability.

Experimental

To obtain stretchability data of conductive ink, the electrical resistance of the conductive ink traces shall be monitored continuously. The four wire Kelvin method is employed to measure the resistance of the conductive ink. An original configuration of the test set up is shown in Figure 1. Four probes are attached to the contact pads of the conductive ink, two probes (I+/I-) are used to apply a constant current and the other two probes (V+/V-) are used to measure the potential drop. The probes can be attached to the contact pads through clips. From our experiment, clips showed relative motion against the contact pads during stretching, causing instability of contact resistance. One alternative option is to use compliant conductive adhesive to glue the probes to the contact pads, however, compliant conductive adhesive may not have adequate strength to withstand repeated stretching cycles. The other option is to use rigid epoxy based conductive adhesive, however, epoxy conductive adhesive typically needs high temperature curing, this may potentially alter the mechanical and electrical characteristics of the conductive ink and substrate, and the rigid-soft interface is prone to cracking during stretching.

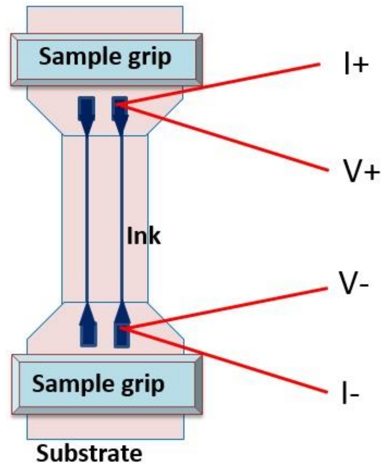

Figure 1: Original stretchability test setup.

The key to address this challenge is to move the contact pads outside the stretching areas, so the contact pads will be free of stresses during stretching. Figure 2 shows a new coupon design to move the contact pads outside the stretch area. Four types of design are embedded in the coupon. For type 1 design, three line widths are designed: 0.5mm, 1mm and 3 mm.

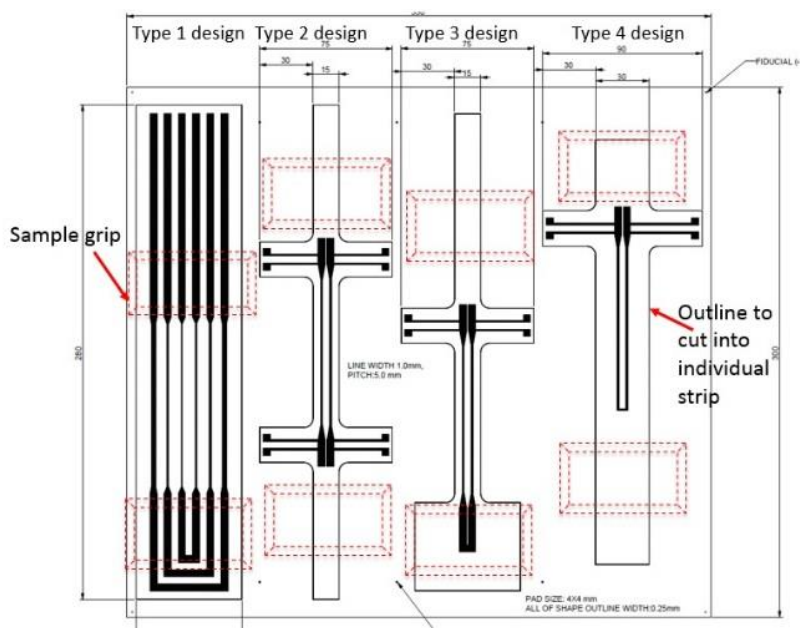

Figure 2: New test coupon design.

A production screen printer platform is used to print stretchable conductive ink on thermoplastic urethane (TPU) substrate. A polyester based screen is made for the experiments with mesh count 305, thread diameter 34 um and thread angle 45 degree. Screen printing parameters are as follows:

- Pressure 14-16 Kg
- Speed: 35mm/second
- Gap: 4mm

After screen printing, the conductive ink is cured at 120ºC for 10 minutes.

Figure 3: Production screen printer.

Figure 4 shows a printed test coupon. TPU substrates from two suppliers are used for this work (Film A: 5 mil thickness and Film B: 4 mil thickness).

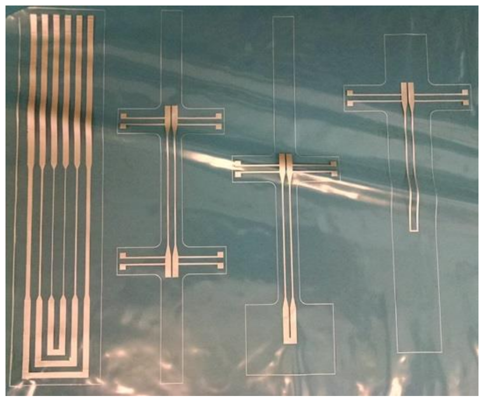

Figure 4: A test coupon made by screen printing.

Figure 5 shows the thickness measurement of the printed ink using a 3D profilometer. The final ink thickness is around 9 um, after curing.

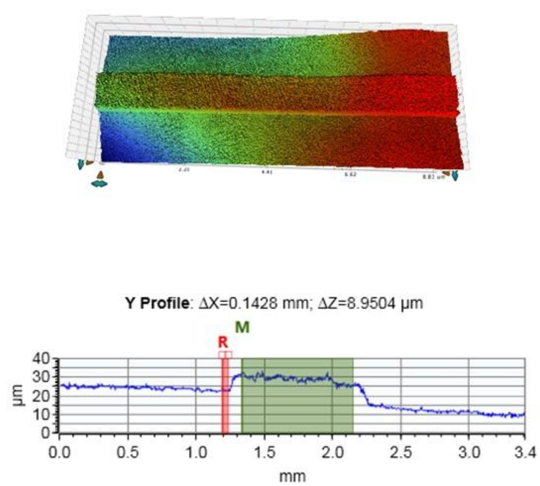

Figure 5: Thickness measurement of the printed ink.

Figure 6 shows the test setup for the stretchability testing. A homemade universal flexibility tester is used for this work [4]. This universal flex tester is designed and made by the company to meet the demands from industry on flexibility testing of wearable electronics. By switching to different motors and fixtures, it can not only perform stretchability testing, but also different types of bending, torsion and rolling. A multimeter is attached to the flexibility tester and synchronized with the tester. The measurement data (resistance, displacement and force) are automatically outputted to the control computer once the test is initiated.

Figure 6: Stretchability test setup.

Results and Discussion

Figure 7 shows the first cycle stretchability curve for the conductive ink. The conductive ink is stretched at a speed of 1mm/second. The initial resistance is around 5 ohms, when it is stretched, the resistance starts to increase, eventually to 164 ohms (peak resistance), a more than 30 times increase, at 50% stretch when the tester stops stretching. After the load is released, the resistance shows a reversal trend, eventually dropping to around 9 ohms (valley resistance) when the experiment is stopped. The electric resistance of the conductive ink is not 100% reversible. A tension set of the TPU substrate is clearly observed, namely, after the load is fully released, the TPU film does not return to its original length, as well as the conductive ink circuitry. Accordingly its final resistance shows a certain increase as compared to the initial value.

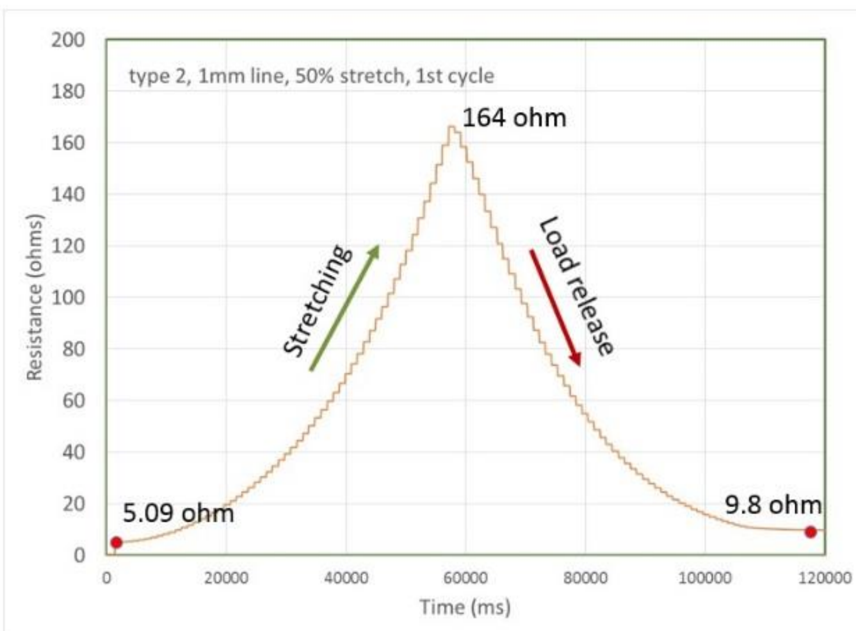

Figure 7: First cycle stretchability.

Figure 8 shows the repeated stretchability cycles for the four designs at a maximum stretchability of 50%. No obvious discrepancies are observed between these four trace designs, with all showing consistent increase in both peak resistance and valley resistance with the latter to a much lesser extent.

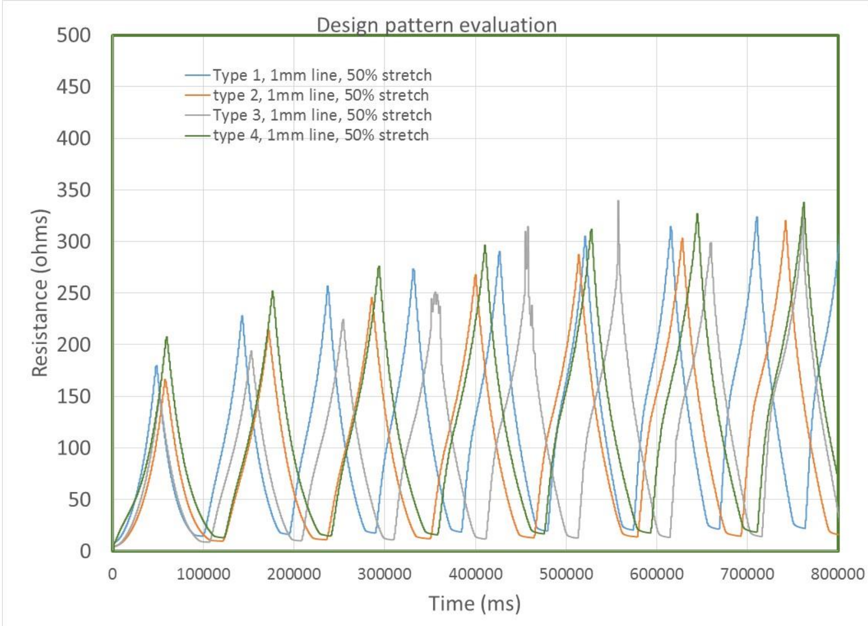

Figure 8: Repeated stretch cycles of conductive ink.

Three test speeds are used to evaluate how the speed affects the ink stretchability: 1mm/s, 5mm/s and 10mm/s. For the stretchable length of 100mm, these speeds correspond to a strain rate of 0.6(mm/mm)/minute, 3(mm/mm)/minute, and 6(mm/mm)/minute respectively. These strain rates are considerably higher than what is recommended for tensile testing by IPC standards, typically in the range of 0.05(mm/mm) to 0.5(mm/mm) per minute [5]. However, in actual applications, the strain rate may be even higher.

Figure 9 shows the hysteresis of first cycle stretchability of the conductive ink at the three test speeds. At the higher speed, a considerable higher resistance is observed. More noticeably, at the speed of 10mm/s, the resistance continues to increase for one or two seconds even if the machine has stopped pulling. The conductive ink is composed of metal flakes, so during stretching the metal flakes slide against each other while maintaining conduction, which will inevitably cause an increase in resistance. At high pulling speed, the metal flakes may have the momentum to move further even when the substrate has stopped stretching, causing continued increase in resistance.

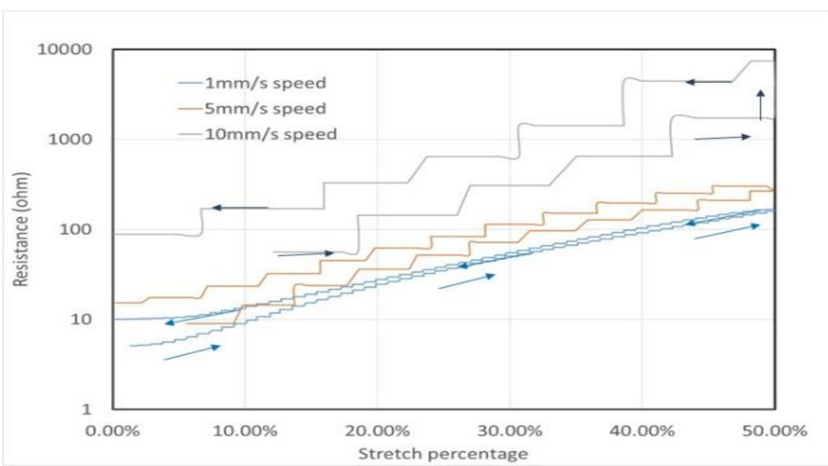

Figure 9: Test speed effect on stretchability.

At high pulling speed the conductive ink shows considerably higher peak resistance with stretching cycles. Figure 10 shows the resistance curves at the above three speeds. Higher pulling speed causes more instability of the resistance of the printed ink.

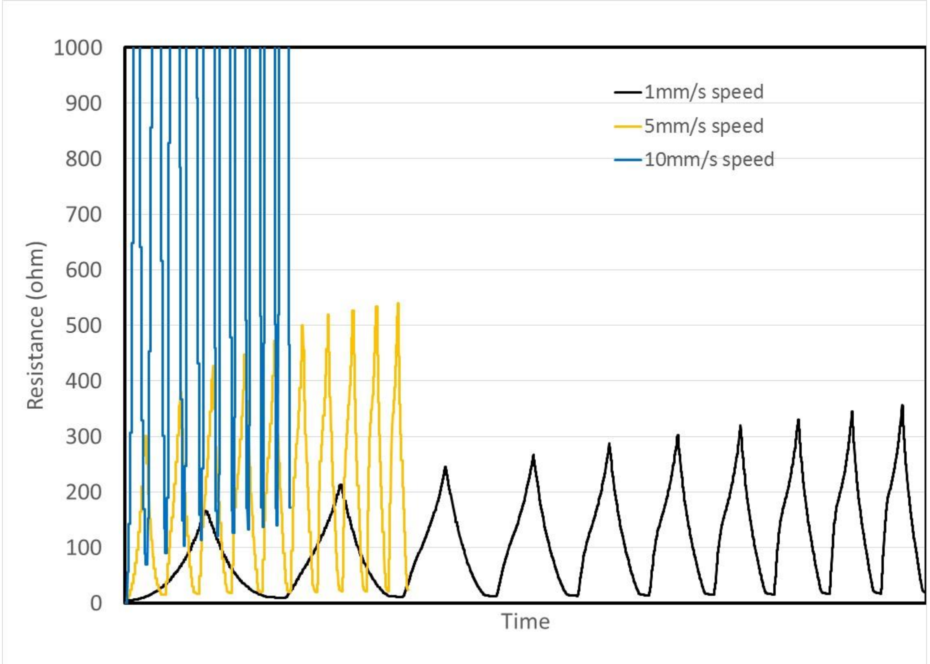

Figure 10: Test speed effect on repeated stretchability

The stretchability of the TPU film substrate will inevitably affect the stretchability of the conductive ink. As mentioned earlier, the tension set of the TPU film may be one major factor for the irreversible resistance recovery of the conductive ink. Although they belong to the same product family, TPU films may have a sharply different composition and manufacturing process, therefore different TPU films may have quite different characteristics.

Figure 11 shows the first cycle stretchability of the conductive ink on two TPU films (Film A and Film B). These two films come from two suppliers, both claim at least 500% stretchability. Figure 12 shows the repeated stretchability for these two films. A significant difference can be observed on the stretchability of the ink printed on these two films. Film A clearly shows better performance as compared to B. Different factors may play a role causing the differences, such as film durometer (hardness), film rigidity, bonding strength with ink, etc. Further studies are needed to understand how these factors affect the ink stretchability and reveal the fundamentals behind.

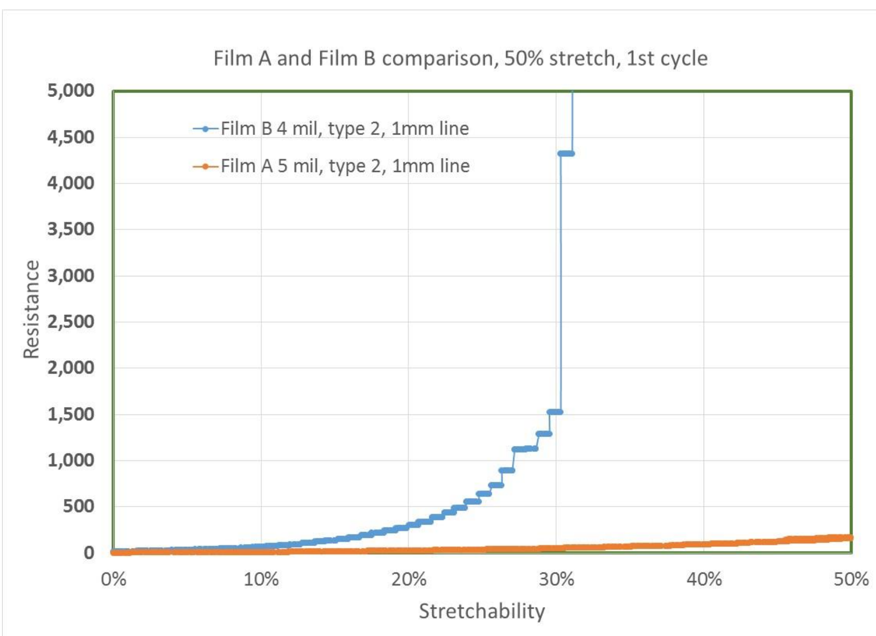

Figure 11: Substrate effect on first cycle stretchability

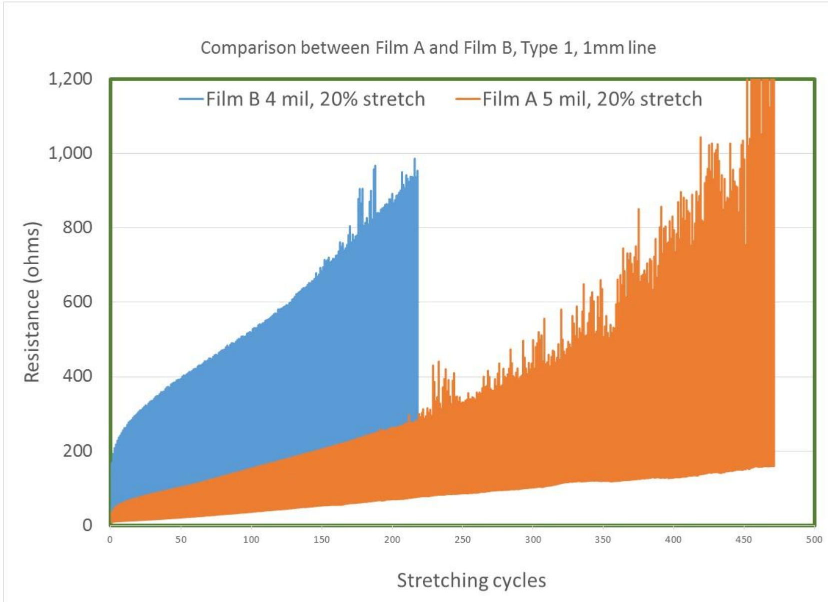

Figure 12: Substrate effect on repeated stretchability.

Figure 13 shows the effect of line width on the ink stretchability. More stable resistance is observed for the larger line width.

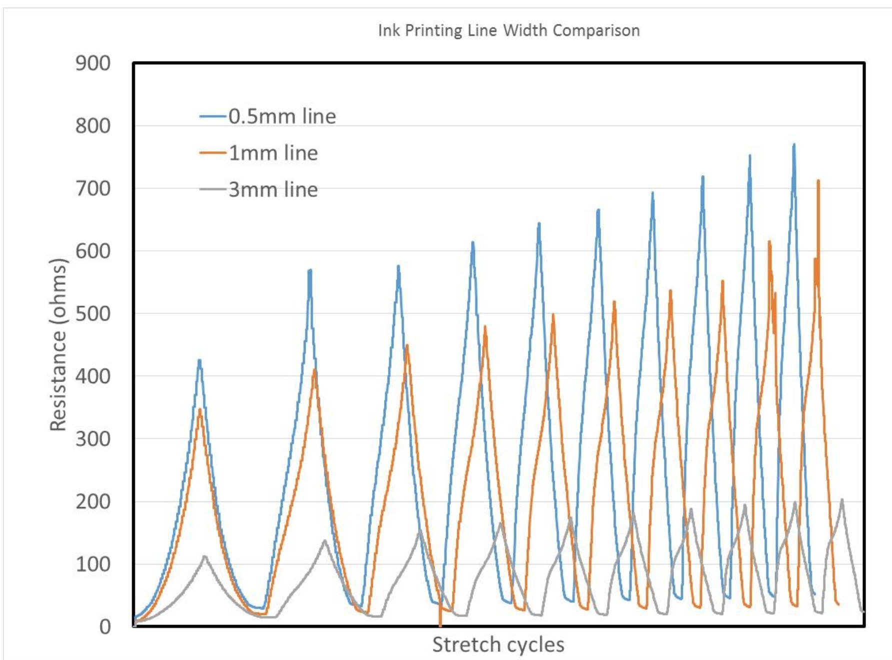

Figure 13: Effect of ink line width on stretchability.

Figure 14 shows the high cycle stretchability of the conductive ink at different stretch percentages (5%, 10%, 15%, 20%, 30% and 50%). As discussed earlier, both peak resistance and valley resistance of the printed ink show continuous increases with stretching cycles. At some point, resistance spikes start to show up, indicating instability of the printed ink circuitry. The metal flakes may loosen up with the polymers holding them with more stretching cycles, causing these resistance spikes.

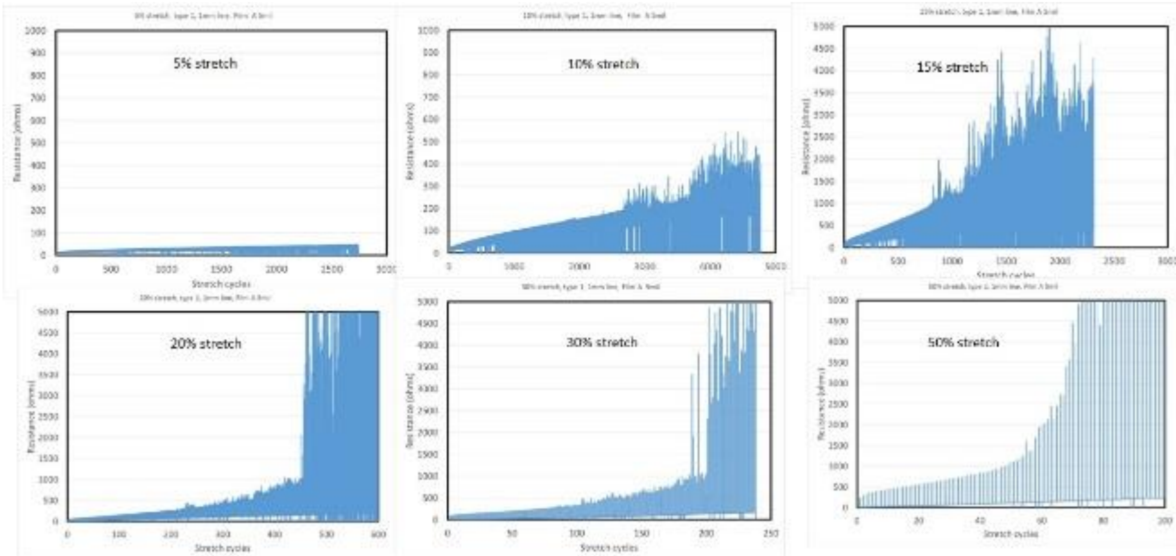

Figure 14: High cycle stretchability of conductive ink.

Figure 15 and Figure 16 show the simulation of peak resistance and valley resistance for 30% and 50% stretch. The peak resistance data are selected prior to resistance spikes. The peak resistance can be fitted very well using an exponential function, while the valley resistance follows a linear relationship with stretching cycles.

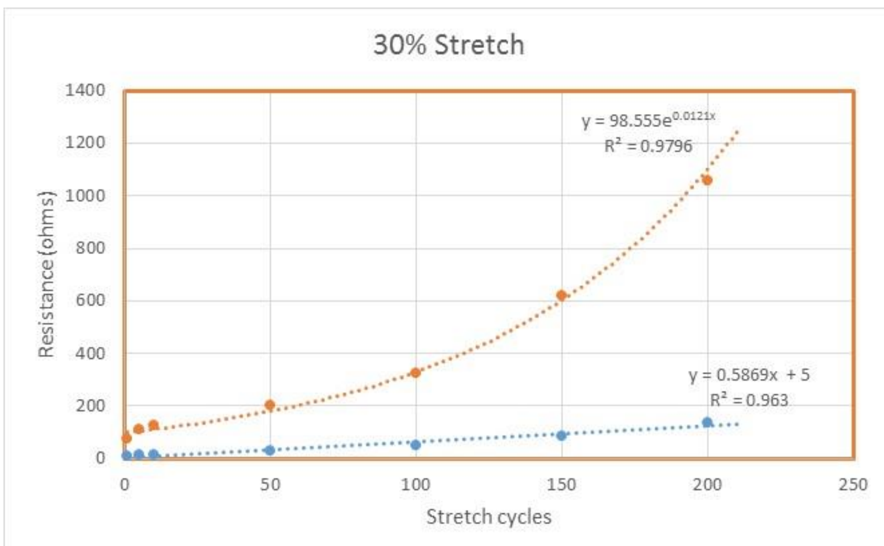

Figure 15: Simulation of peak and valley resistances for 30% stretch.

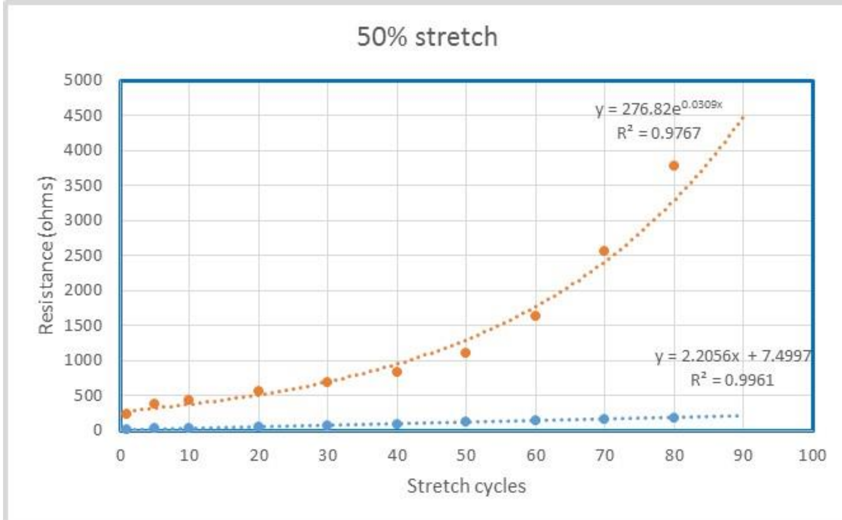

Figure 16: Simulation of peak and valley resistances for 50% stretch.

Figure 17 shows the stretchability cycles versus stretch percentage for the stretchable ink studied. More than 2,500 cycles can be achieved for 10% stretch, close to 500 cycles for 20% stretch.

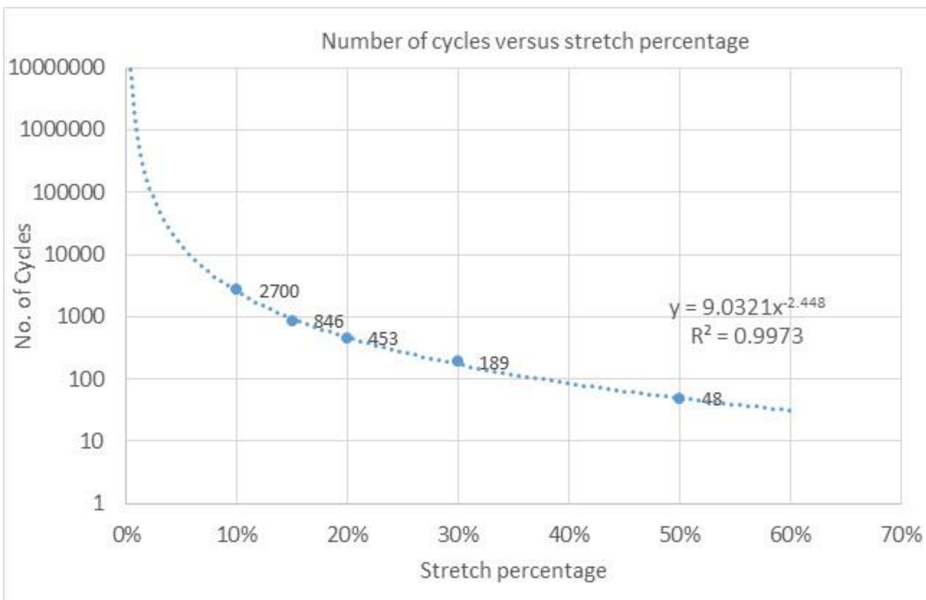

Figure 17: Stretchability cycles versus stretch percentage.

In order to optimize the stretchability of the printed ink, we need to understand the mechanisms behind stretchability. SEM inspection on the conductive ink printed on the two substrates Film A and Film B is performed while the printed ink is kept under a constant stretching state (both 50% stretch and 100% stretch). Figure 17 shows the surface macrostructure of the printed ink at 50% stretch. Ink printed on Film B shows prolonged cracks across the printed ink traces, while the ink printed on Film A only shows minor cracks and most of the metal flakes are still connected to each other. In order to magnify the phenomenon further, the ink is stretched to 100%. Figure 18 show the SEM inspection results at the 100% stretch. Ink printed on Film B clearly shows debonding of the ink to the substrate and large cracks across the whole printed surface. Ink printed on Film A shows more obvious cracks as compared to the ink at 50% stretch, however, the metal flakes are still connected at the peripherals of cracks. Electrical measurement shows the circuitry has become open for the ink printed on Film B at 100% stretch, while the ink printed on Film A still shows conductance even at 100% stretch. This SEM analysis confirms our previous test data showing the ink printed on Film A performs better than the ink printed on Film B. Further studies are needed to evaluate different characteristics of the film substrate and how they impact the ink stretchability, such as roughness, bonding strength, rigidity, tensile modulus, thickness, tension set, etc.

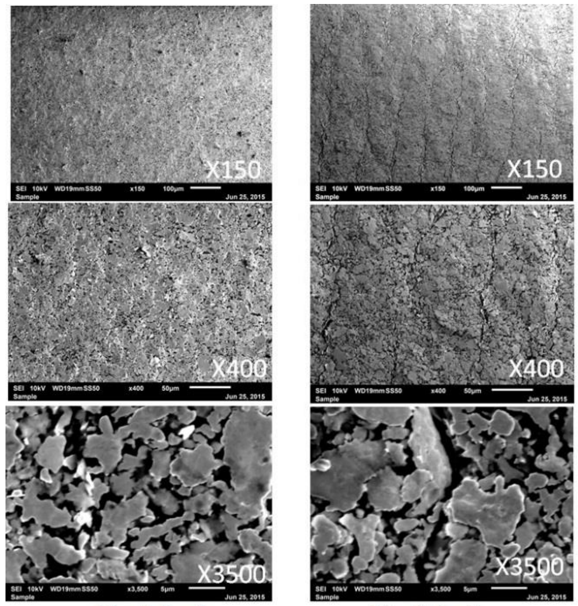

Figure 17: SEM analysis of the printed ink at 50% stretch.

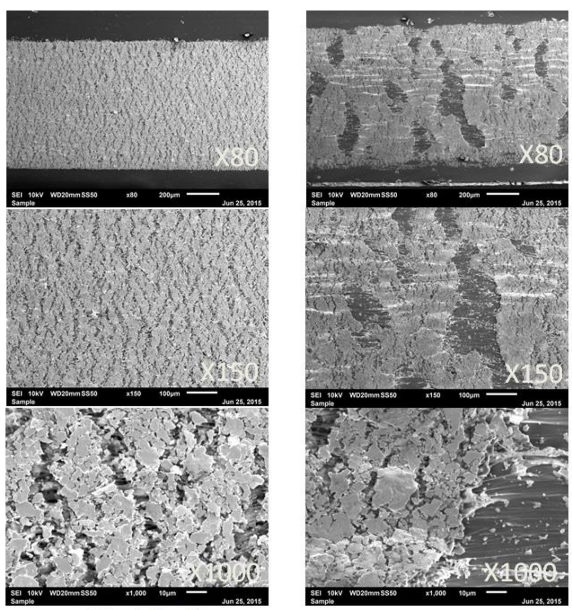

Figure 18: SEM analysis of the printed ink at 100% stretch.

From the SEM analysis, it shows two different stretching mechanisms. The stretchable conductive ink is typically composed of metal flakes embedded in a polymer matrix. At a low stretch percentage, the metal flakes tend to slide against each other, while this may cause an increase in resistance, it maintains the connectivity within the circuitry. When the stretch percentage is high, the polymer matrix may not be able to hold the particles, so cracks will form. The cracks serve as a stress reliever, with metal flakes are still connected at the peripherals of the cracks. With these stress relieving cracks, the printed ink can be stretched at a higher percentage without losing connectivity. This mechanism occurs on the condition that the bonding strength between the printed ink and substrate is adequate to withstand the stresses during stretching. If the bonding strength is low, the metal flakes may break from constraints of the substrate film, causing delamination of the ink from the substrate and accordingly electrical discontinuity.

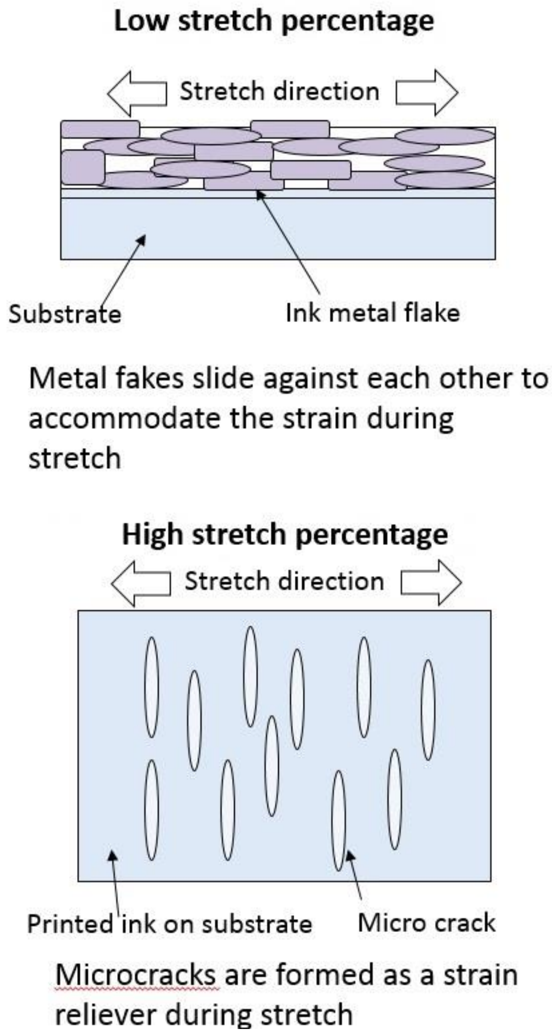

Figure 19: Mechanisms to enable stretchability of conductive ink.

Summary and Conclusions

Extensive experimental work has been done to understand the behavior of a conductive ink under stretching. Standard test coupon is designed and validated to facilitate stretchability testing and ensure test repeatability and consistency. Different design and test parameters are evaluated to understand how they affect ink stretchability.

Test speed (strain rate) clearly affects the ink resistance stability. Higher speed causes a sharper increase in resistance and resistance instability. Substrate material plays an important role in ink stretchability. Further study is needed to further understand how different characteristics of substrate materials affect ink stretchability. For the high cycle stretchability, the ink peak resistance increase typically follows an exponential function with stretch cycles, however, at a certain stretch cycle, resistance spikes show up, indicating internal structural instability of printed conductive ink.

With initial understanding of the printed ink stretchability, the plan is now in place to shed deeper insight on ink stretchability. This includes more extensive work to quantify how different factors affect stretchability, such as dwell time, ink thickness, ink overcoating, ink encapsulation. DoE experiments will be performed to understand interactions between these factors.

This work is in part to support the standardization effort of the IPC D65 subcommittee on ink stretchability. The initial draft on the printed ink stretchability test method is now under review.

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FLEX International

Milpitas, CA

Outline

- Introduction
- Evaluation plan and goals
- Experimental test setup
- Results and analysis
- Summary and conclusions
- Future work

Stretchable Electronics

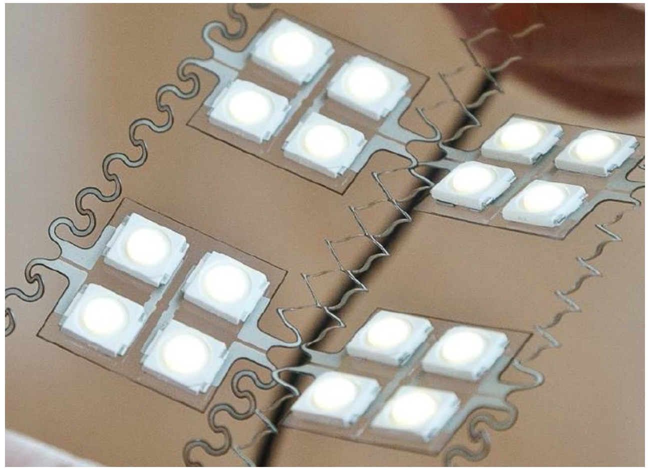
Stretchable electronics concerns electrical and electronic circuits and combinations of these that are elastically or inelastically stretchable by more than a few percent while retaining functionality*

Stretchable electronics has become a popular subject for study and application, along with wearable electronics, since it attempts to mimicry human skin and body, in being stretchable, whilst retaining full functionality

*Source: IDTechEx, Stretchable Electronics and Electrics 2015-2025: Technologies, Markets, Forecasts, 2015

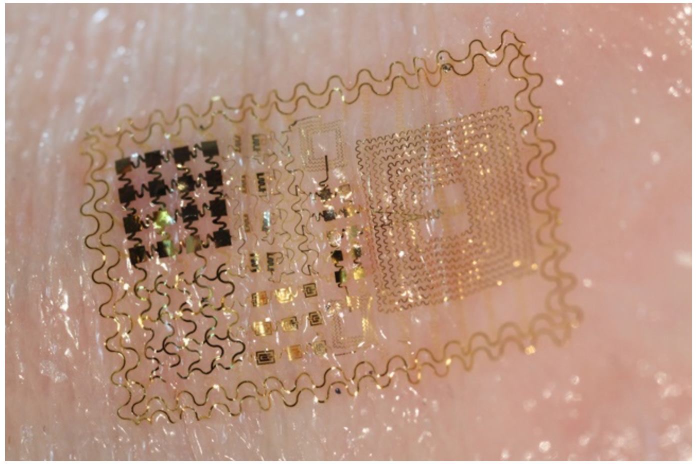
Applications of Stretchable Electronics

Baby sleep monitor (Source: Mimo)

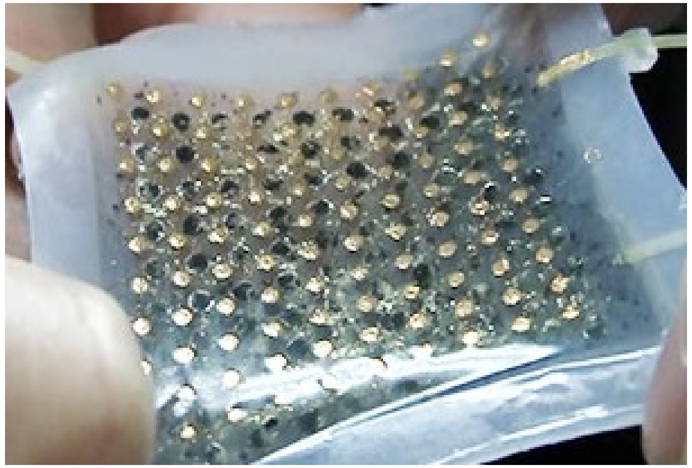


Stretchable LED array (Source: Holst)

Stretchable ECG Sensor laminated to compression shirt (Source: Company)



Stretchable skin patch (Source: MC10)

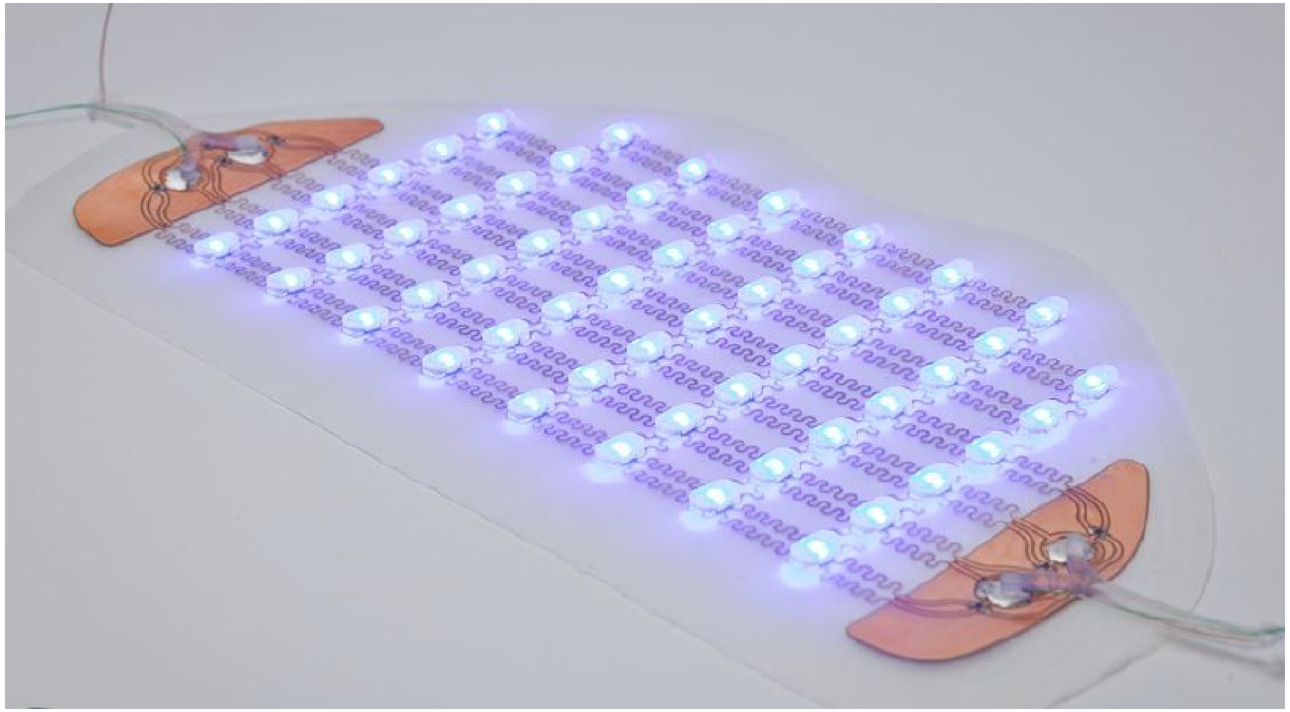


Stretchable battery (Source: UNW)

Key Elements of Stretchable Electronics

The core technologies of stretchable electronics lie in three respects:

- **Stretchable interconnect**
- Stretchable substrate
- Stretchable encapsulation

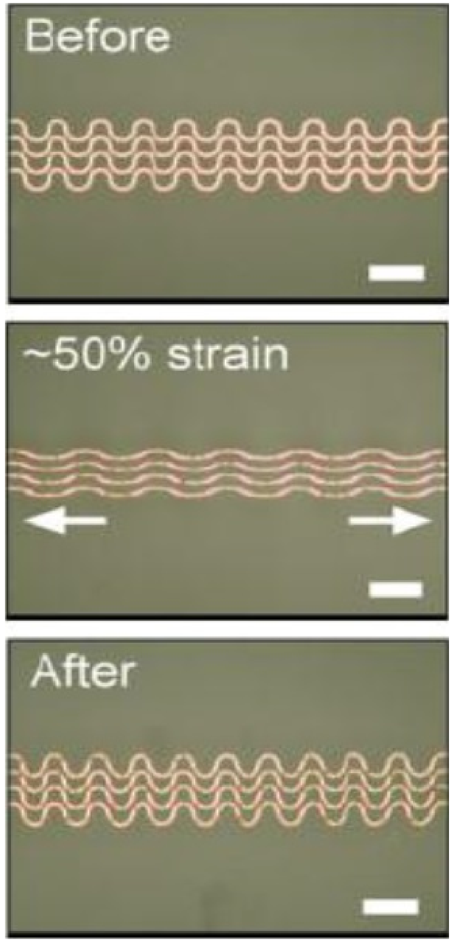


Stretchable Interconnect

- To make interconnect stretchable, the traditional way is to make the line a meander or serpentine shape, to serve as a stress relieve
  - This can be solid wire or printed ink
  - However, the serpentine shape takes extra space, either x y plane or z direction

- A new way to make stretchable circuitry is to directly print stretchable ink in straight lines on stretchable substrate
  - The ink is more stretchable in serpentine shape
  - The ink is screen printable, either flatbed or roll to roll (R2R), enabling high volume production

- However, few data available on the ink stretchability
  - Need to develop, validate and standardize stretchability test method
  - Need to understand how different factors affect stretchability of printed ink



Meander shaped interconnect (Courtesy of IMEC)

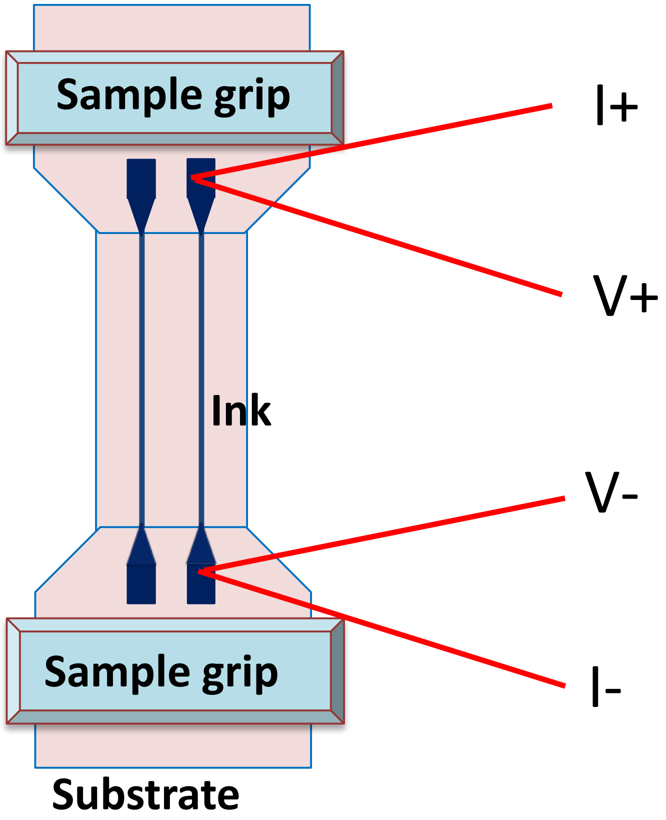
Stretchable Ink Evaluation

Project Goals:

- Obtain a better understanding of the behavior of printed conductive ink during stretching
- Validate test coupon design for stretchability study
- Obtain optimal design and test parameters to optimize stretchability and result consistency
- Support IPC D65 standardization on stretchability testing of printed ink

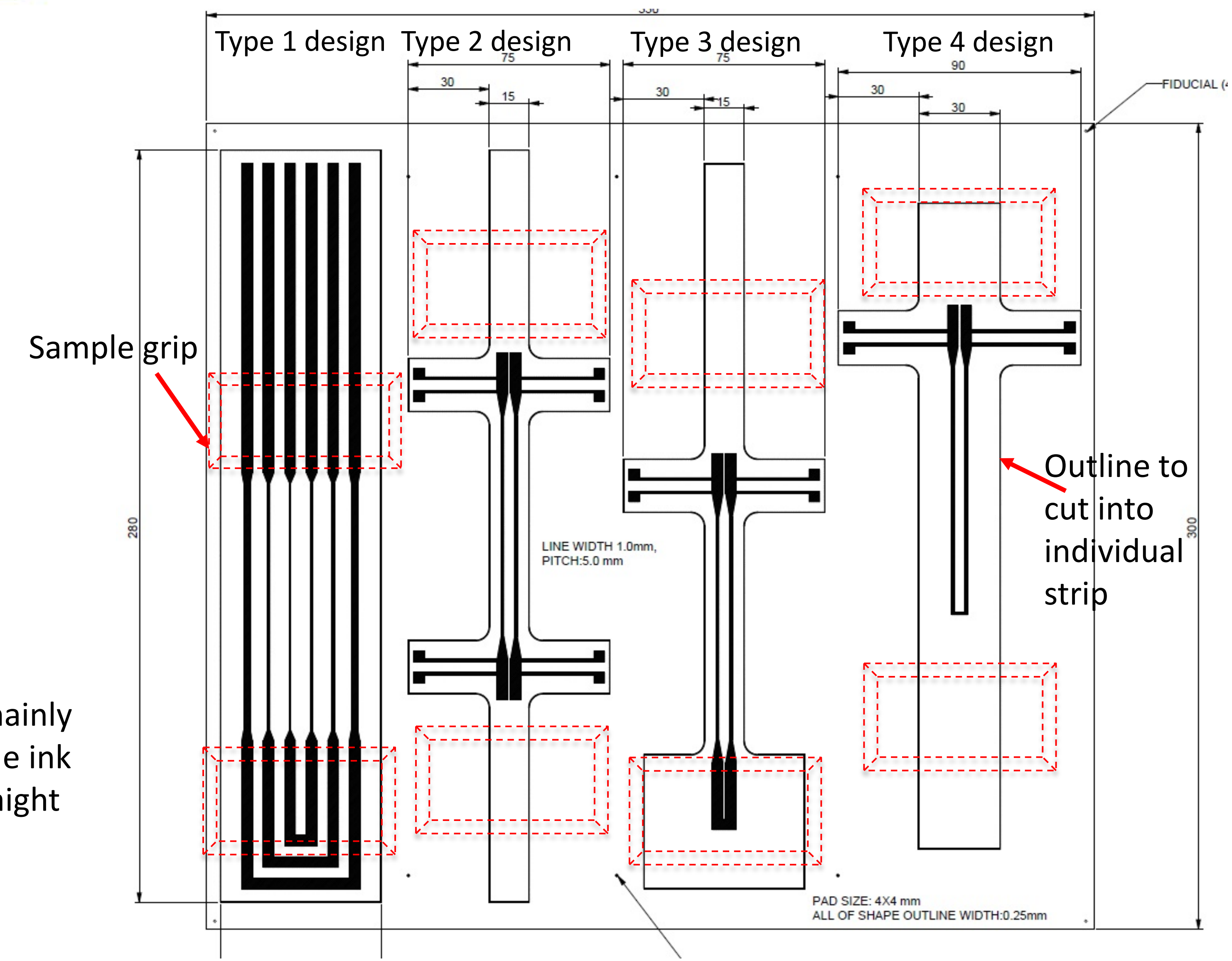
Issues with Current Test Setup

Conventional way of probe connection (Contact pads within stretched area)



- Resistance shall be monitored during stretchability testing
- Issues with conventional connection:
  - Clip may move during stretching
  - Rigid epoxy conductive adhesive may need high temperature curing and the rigid-soft interface is prone to crack during stretch
  - Compliant conductive adhesive may not have adequate strength to withstand stretch cycles
  - All these leads to instability of resistance measurement, complicating resistance measurement of conductive ink
- The key to address this is to move the contact pads outside the stretched area, so the contact pads will be free of stresses during stretching

Test Coupon Design

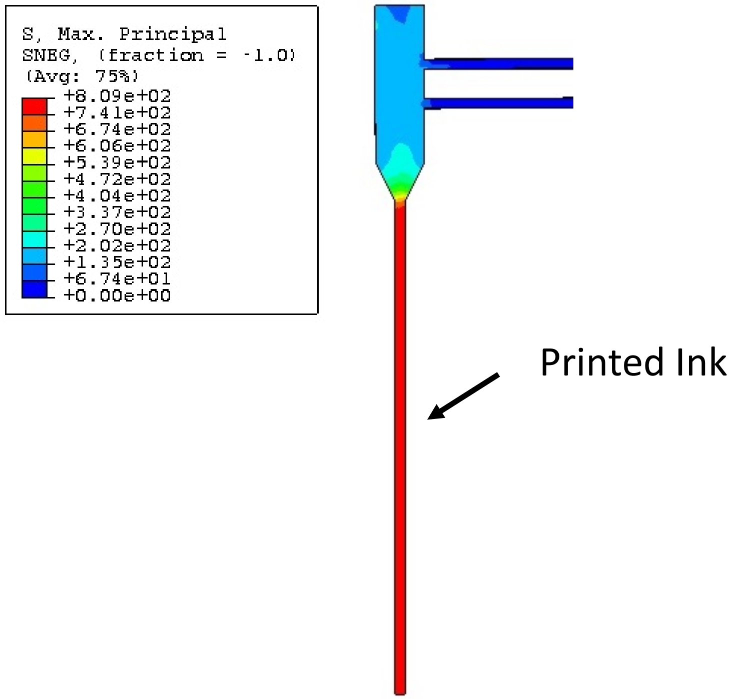


- Design types
  - Type 1
  - Type 2
  - Type 3
  - Type 4
- Line width
  - Type 1(0.5mm, 1mm and 3mm)
  - Type 2, 3, 4 (1mm)

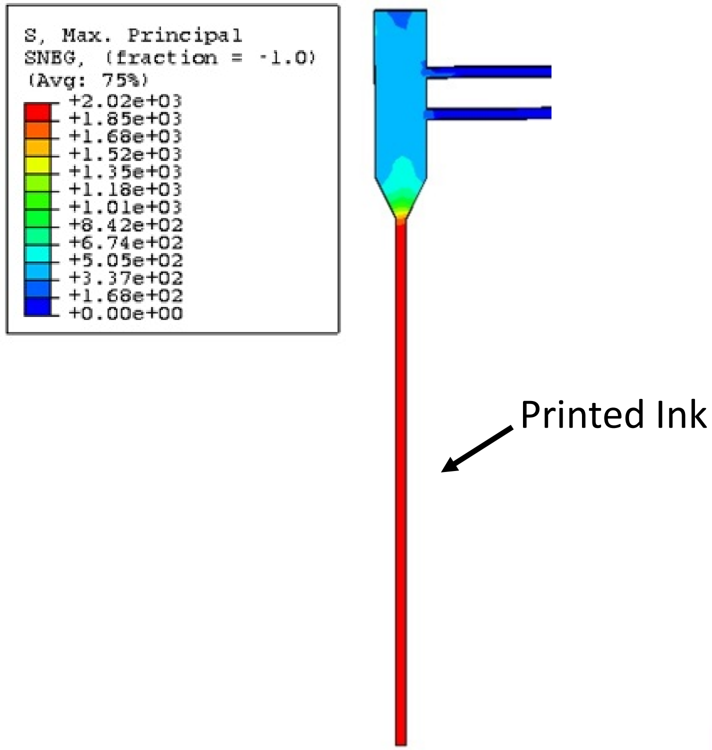
Note: This design is mainly to evaluate stretchable ink materials, so only straight lines are designed.

Stress Simulation During Stretch (Type 2)

20% Stretch



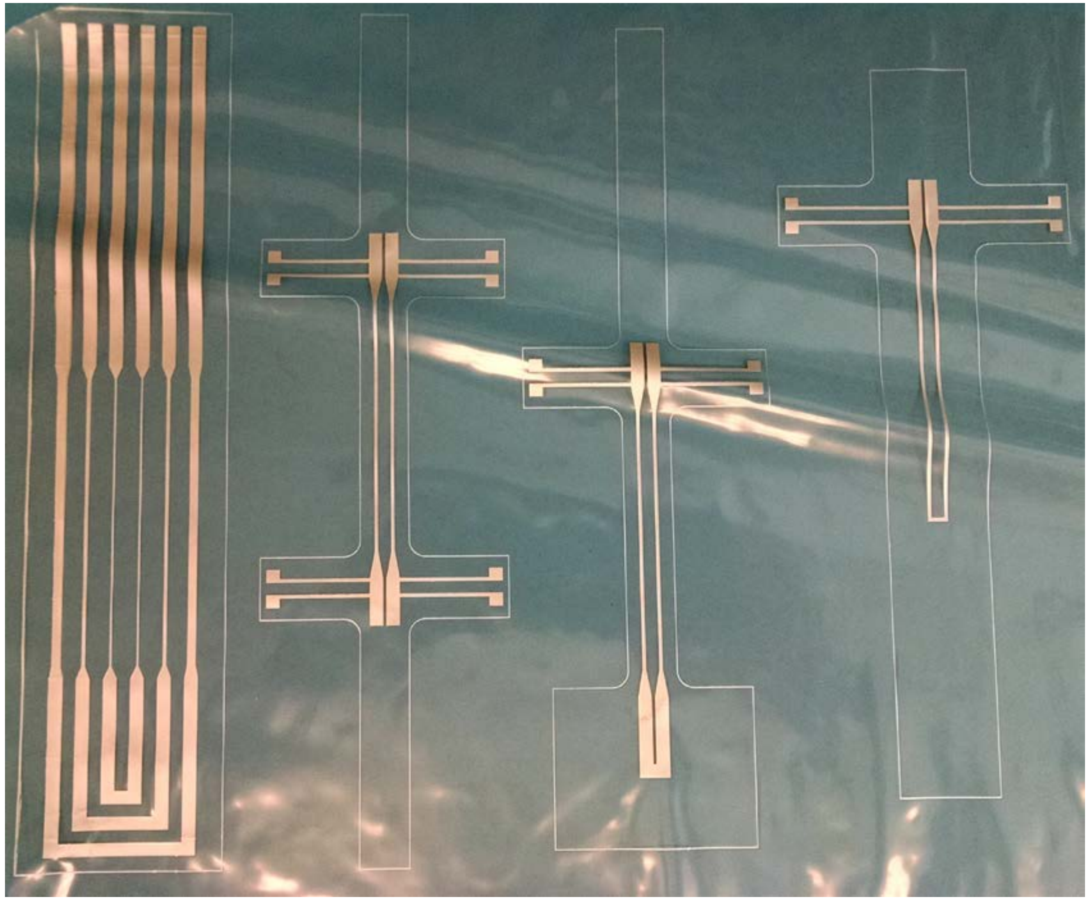
50% Stretch

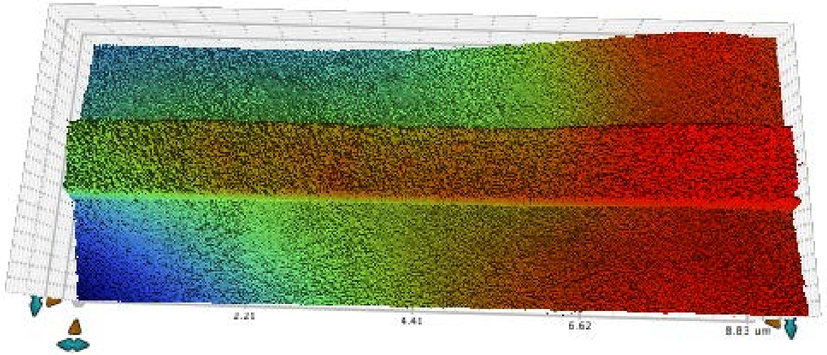


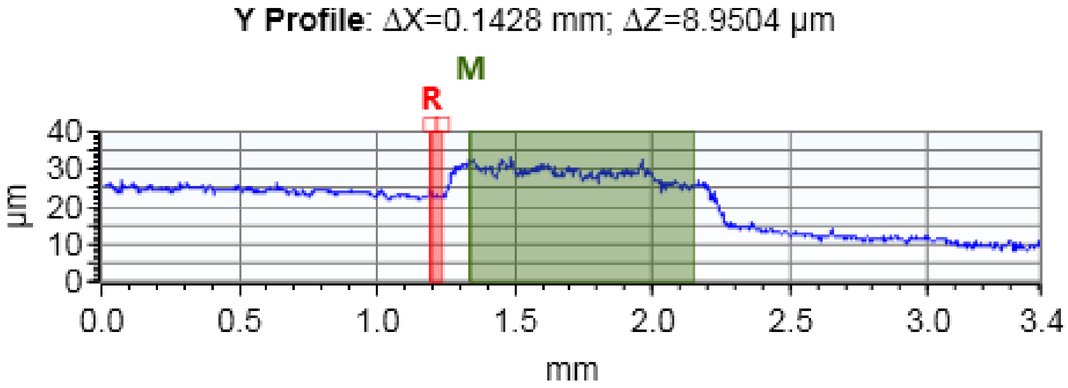
Stretchable Ink Printing

- Screen
  - Polyester
  - Mesh count 305 (by inch)
  - Thread diameter: 34 um
  - Angel: 45°
  - Emulsion thickness: 10 um
- Printing parameters
  - Pressure 14-16 Kg
  - Speed: 35mm/s
  - Gap: 4mm
  - Double printing
- Substrate: Thermoplastic polyurethane (TPU) film
  - Film A: 2 mil and 5 mil
  - Film B: 2 mil and 4 mil
- Curing
  - 120^{o}C for 10 minutes

Printed Samples (after curing)

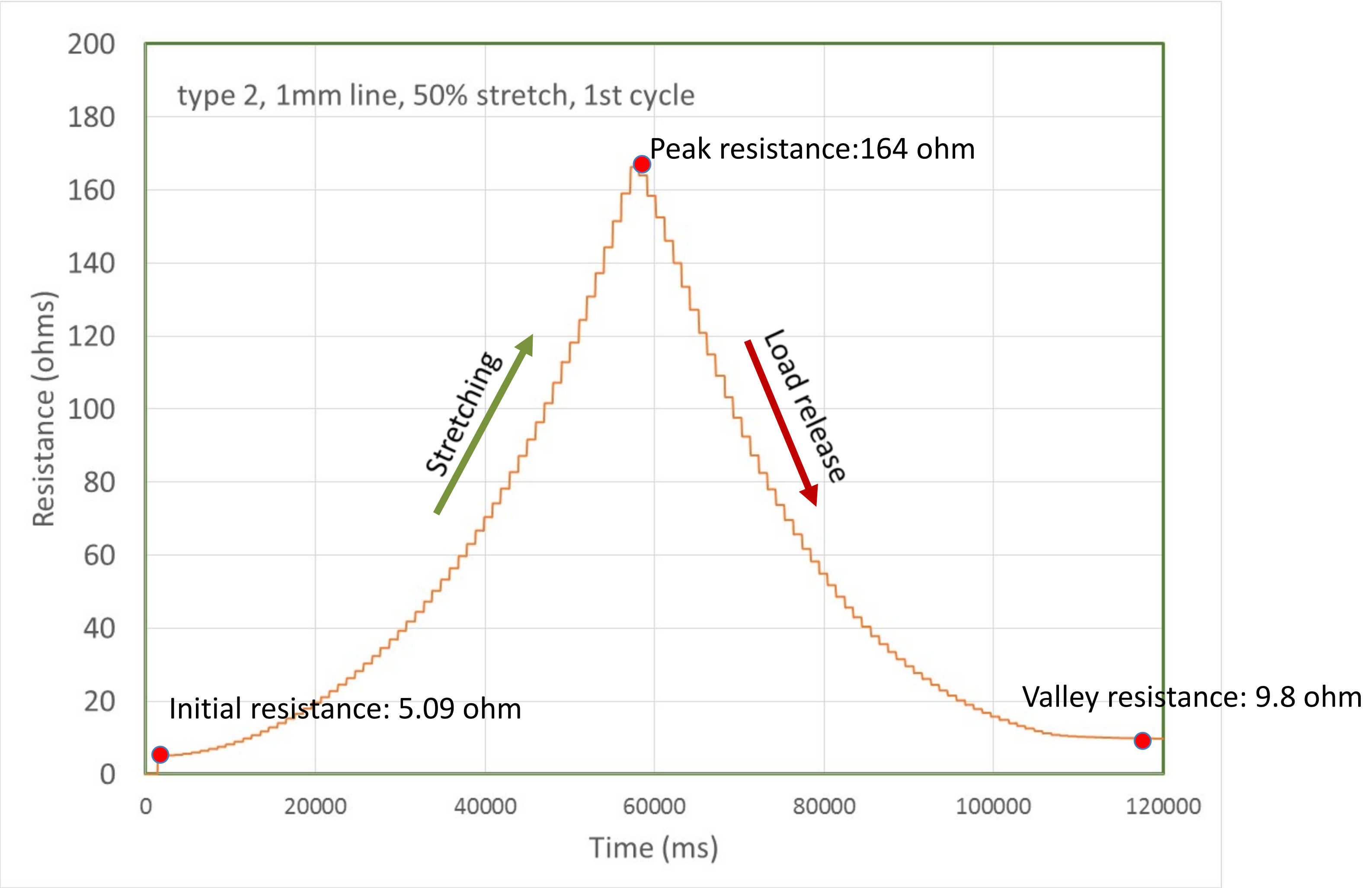




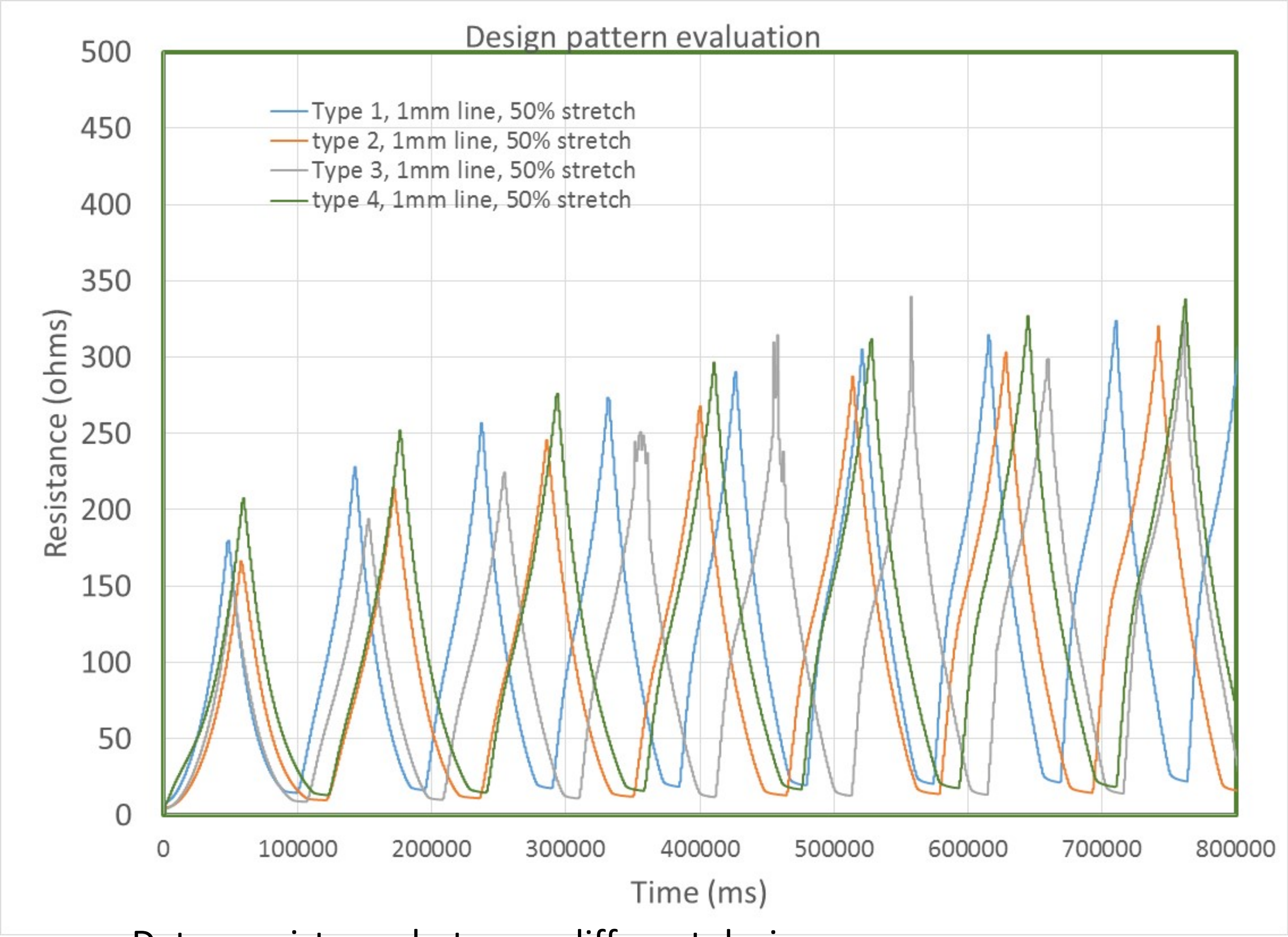


Stretchability Testing Machine

Typical First Cycle Stretchability

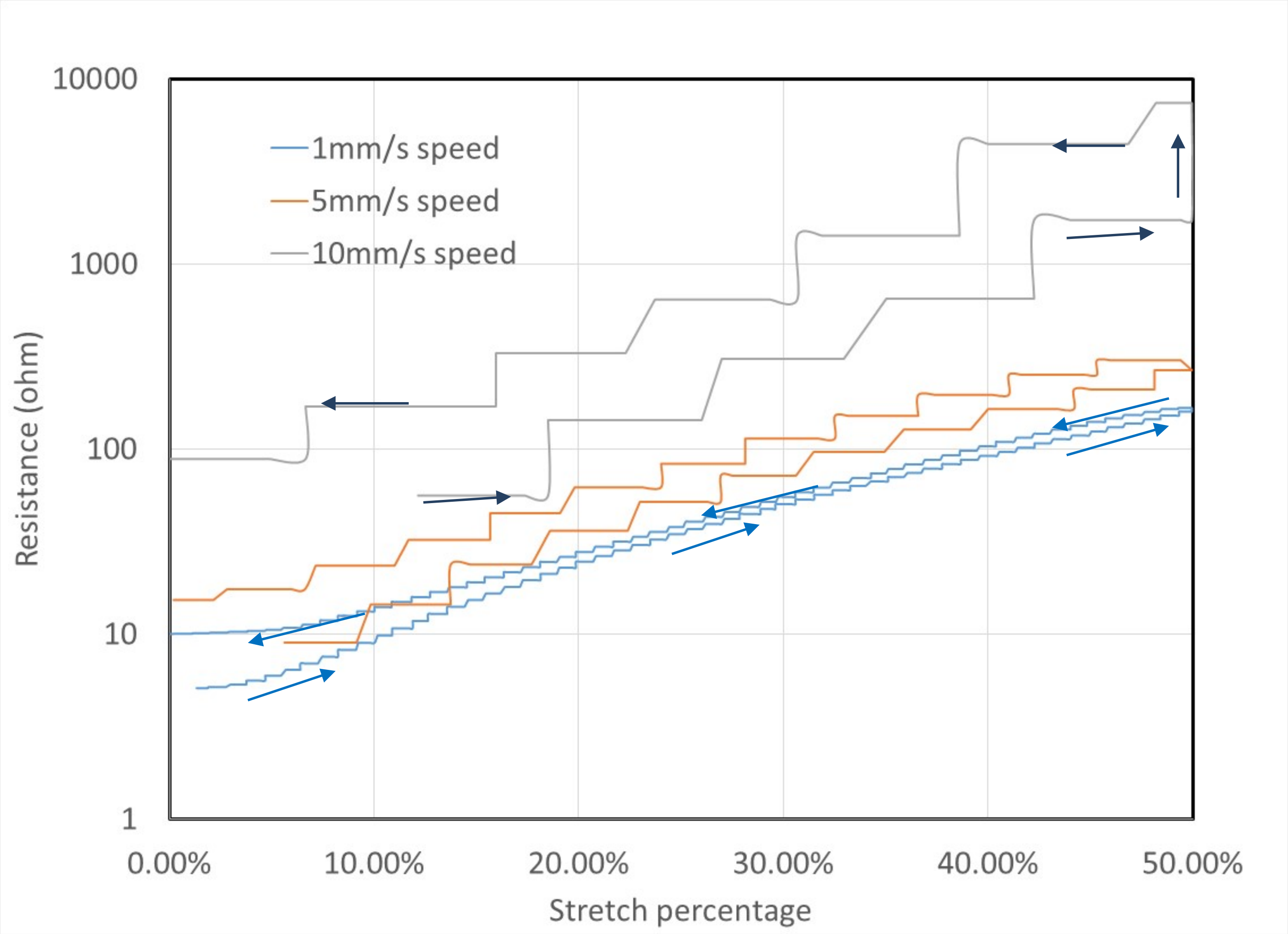


Cyclic Stretch Behavior of Ink



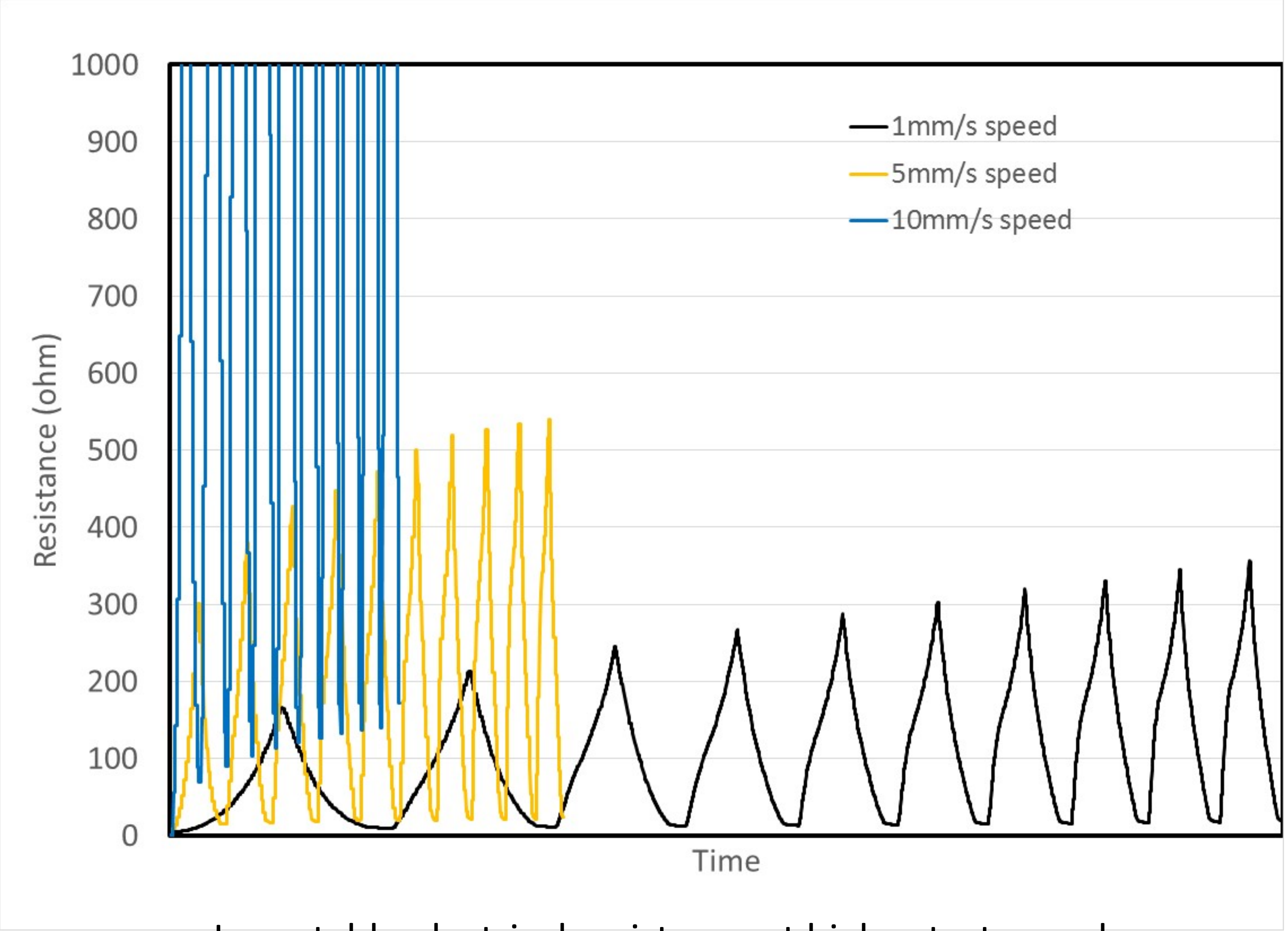
- Data consistency between different designs

Test Speed Effect



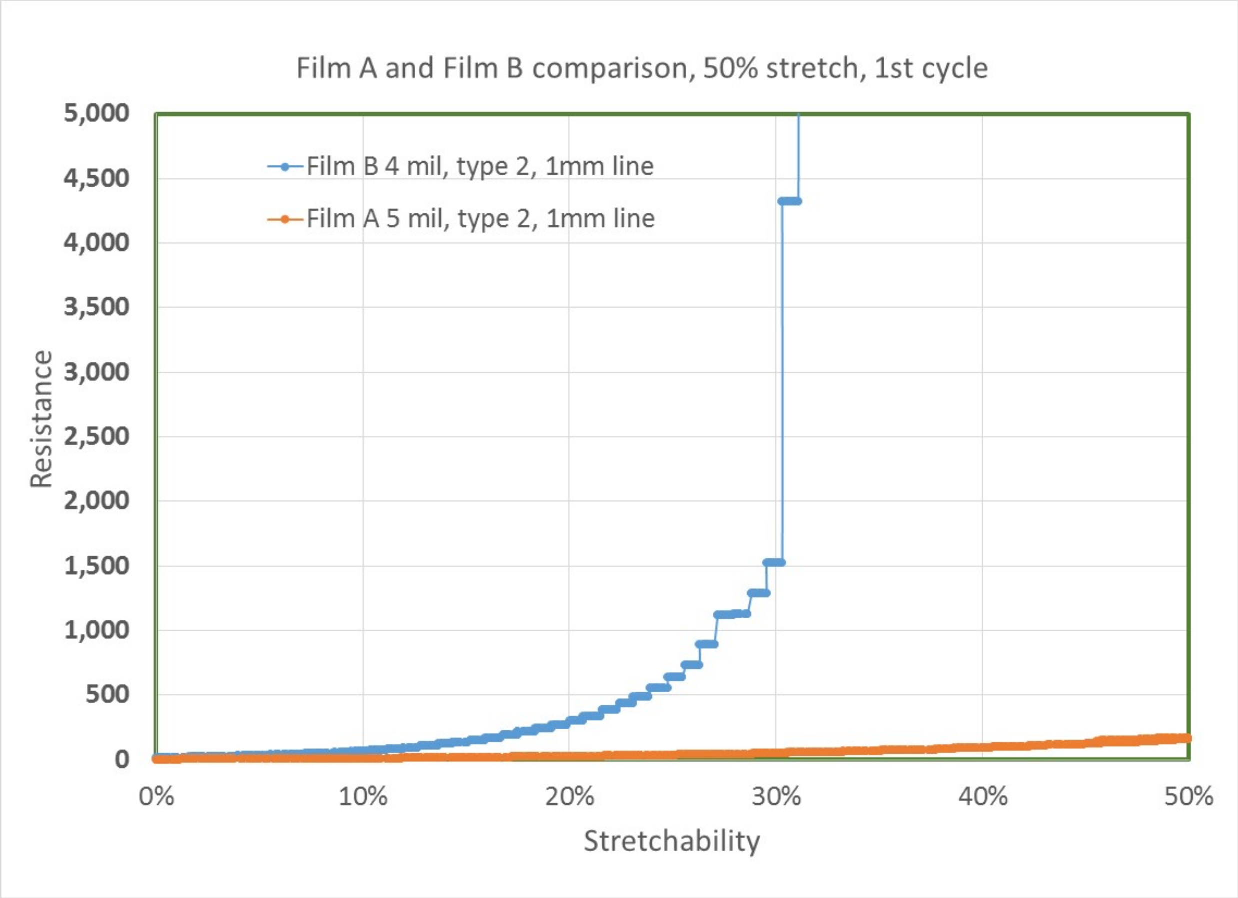
- Sharper increase in electrical resistance at higher test speed
- More obvious hysteresis of electrical resistance at higher test speed

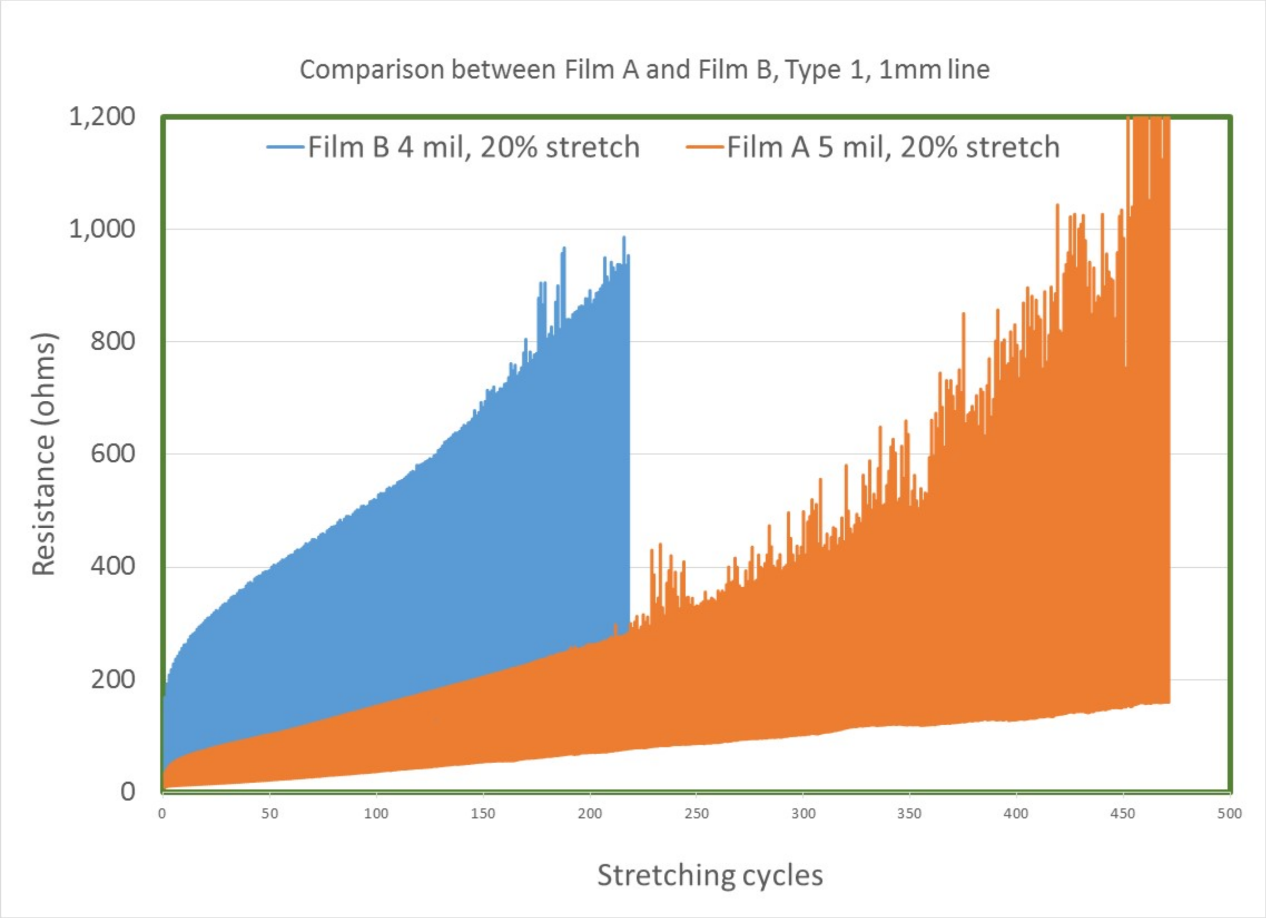
Test Speed Effect (Cyclic Stretchability)



- Less stable electrical resistance at higher test speed

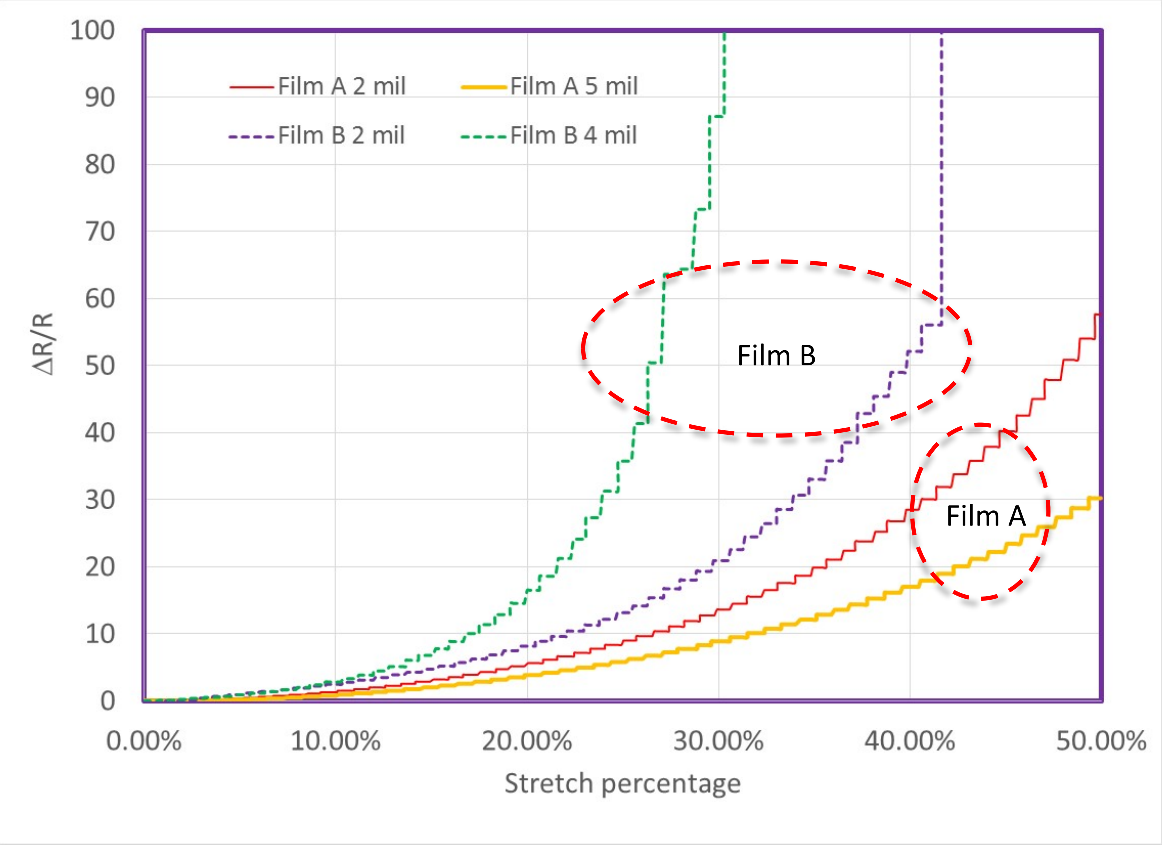
Substrate Material (TPU) Effect

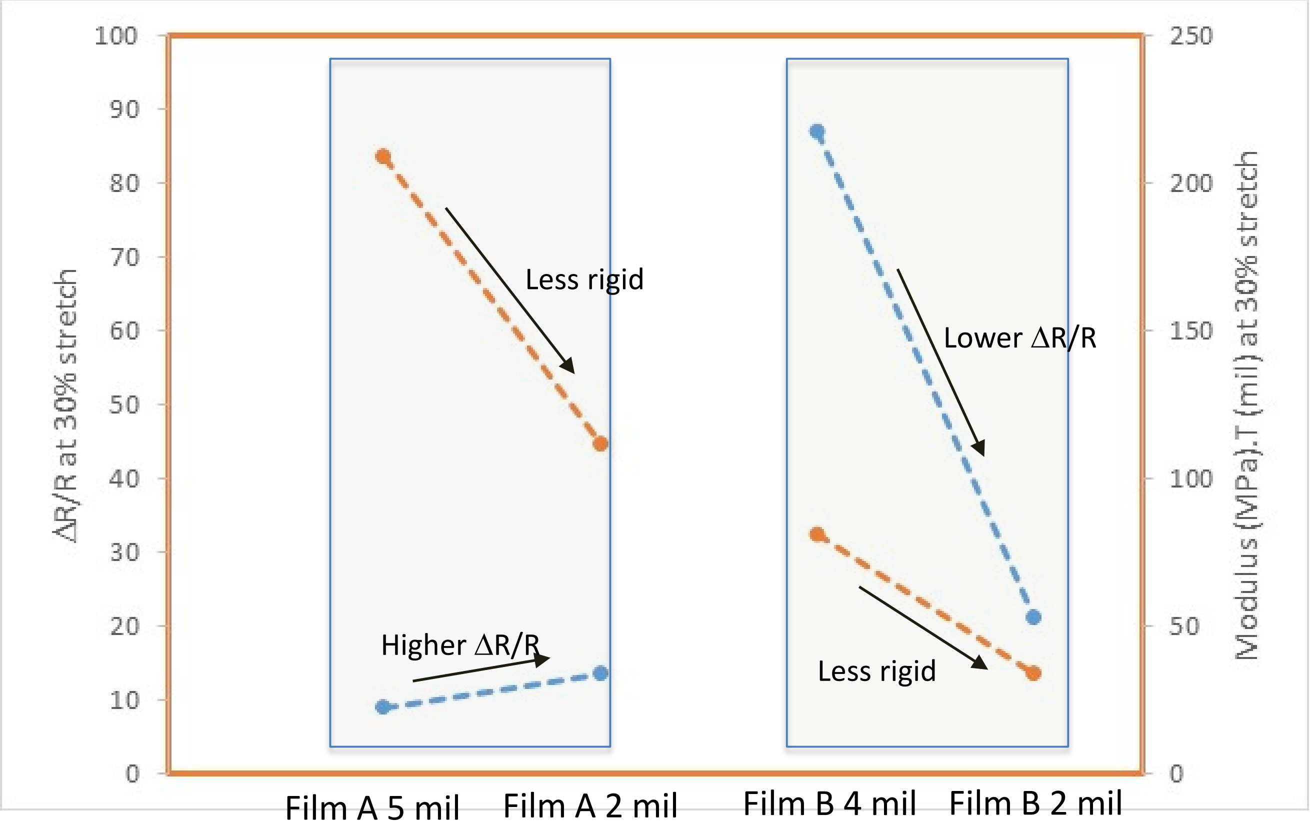




- More stable electrical resistance observed for film A as compared to film B

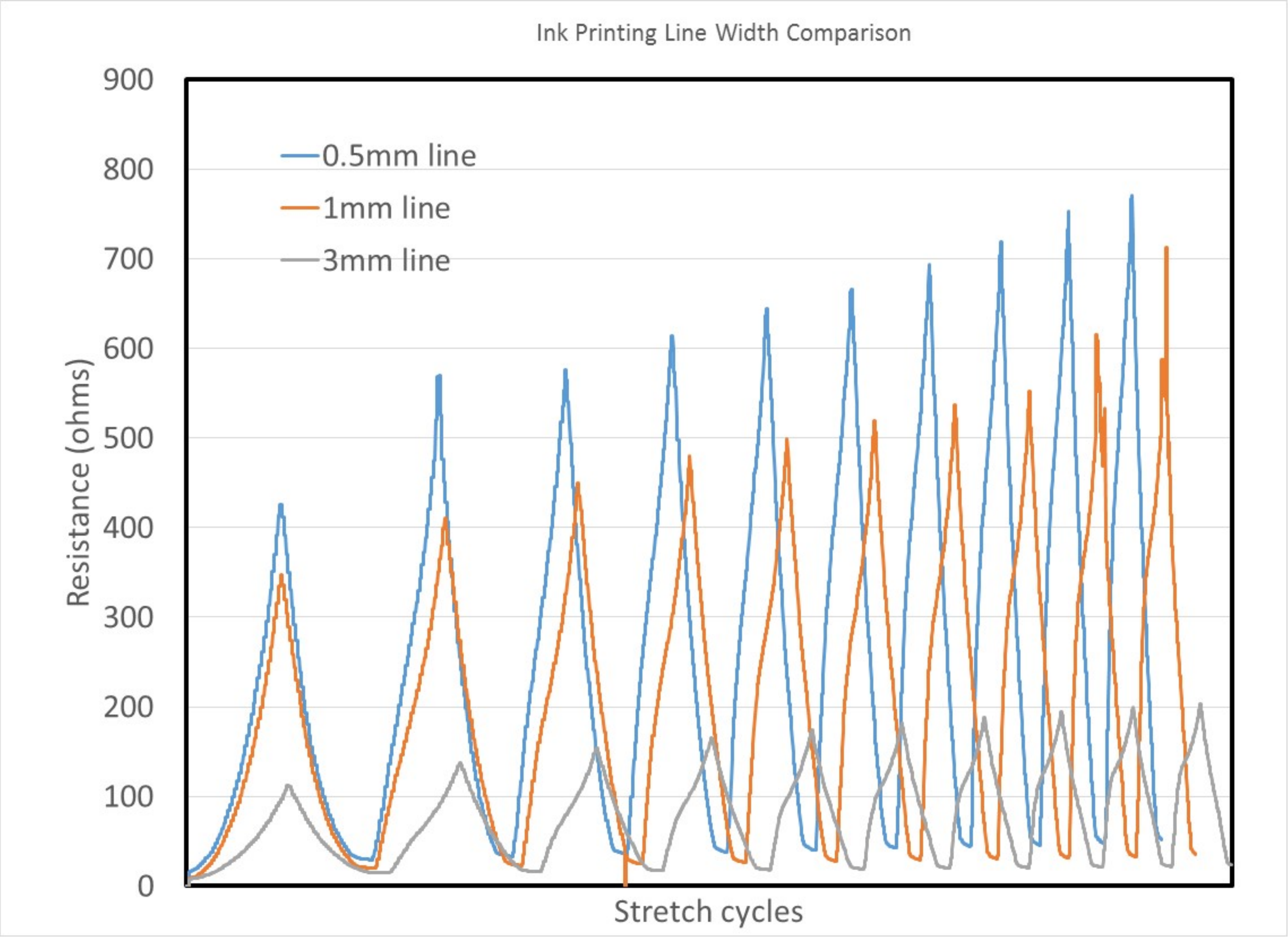
Substrate Material/Thickness Effect





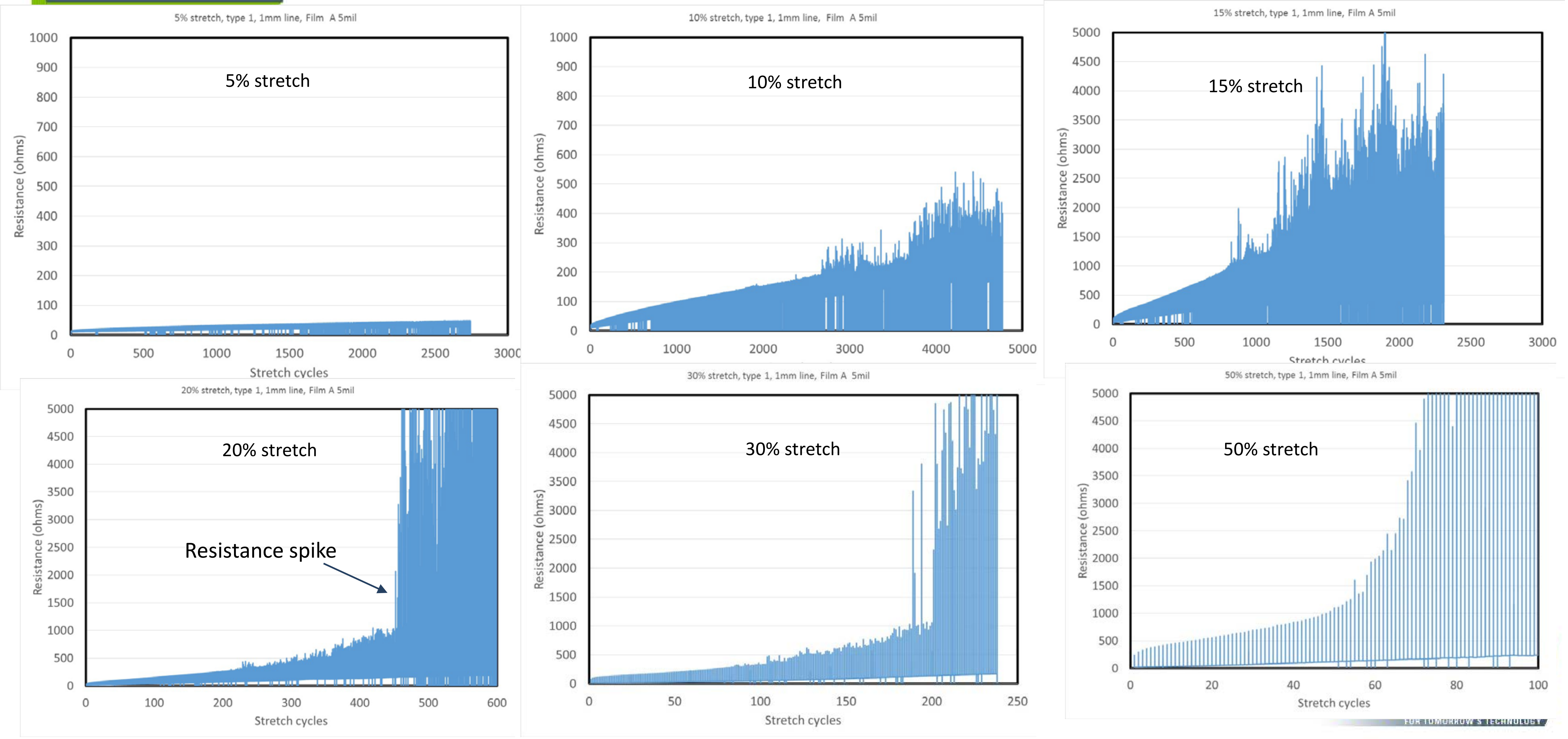
- Profound effect of substrate material and thickness
    - Film A (more rigid): thicker film performs better
    - Film B (less rigid): thinner film performs better

Ink Line Width Effect

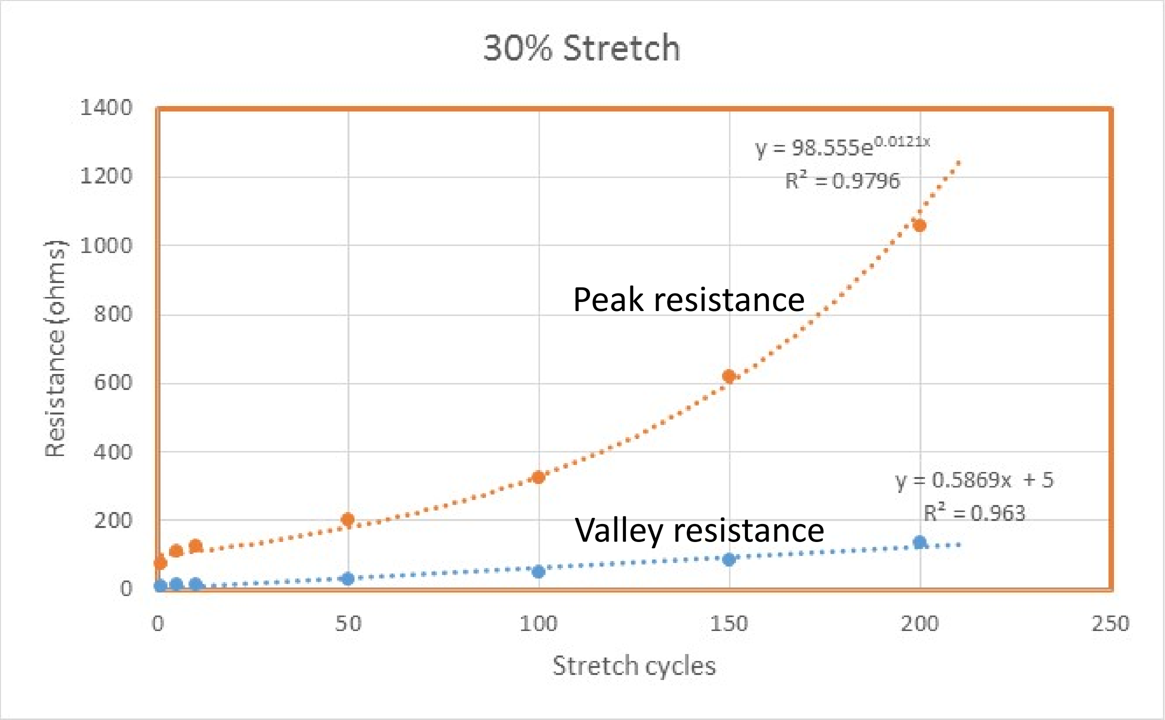


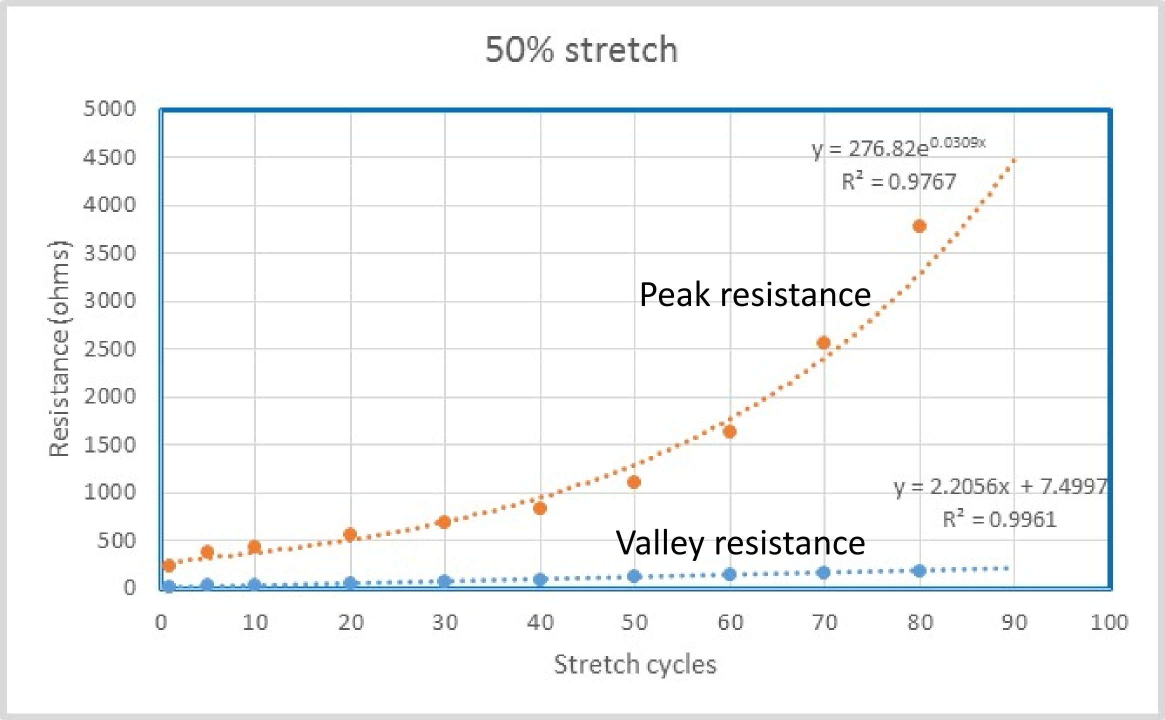
- More stable resistance for wider printed ink lines

Cyclic Stretchability



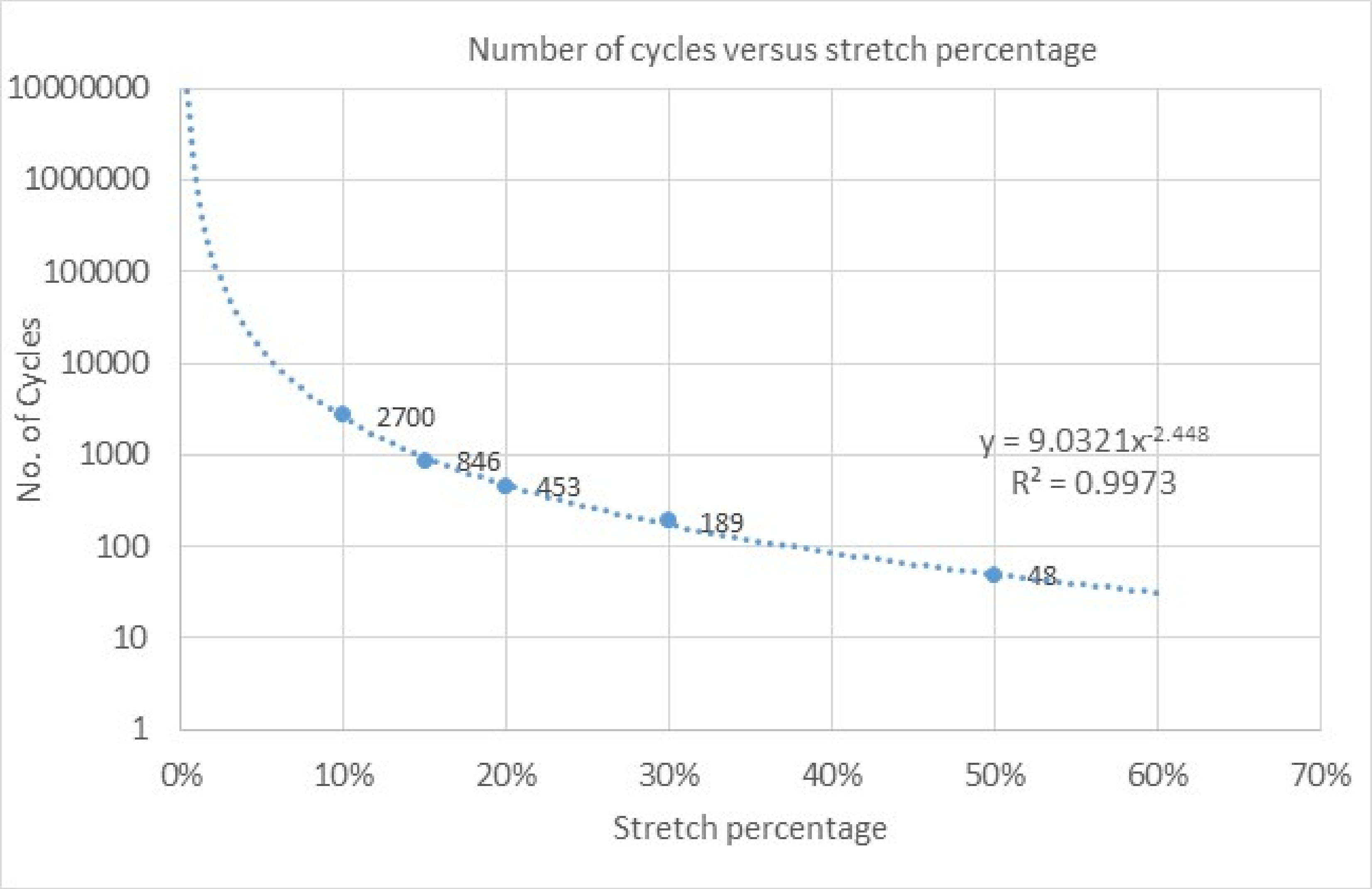
Simulation of Cyclic Stretchability





- Exponential function can be used to fit the trend of peak resistance of the printed ink during stretching, while the valley resistance follows the linear relationship with stretch cycles.

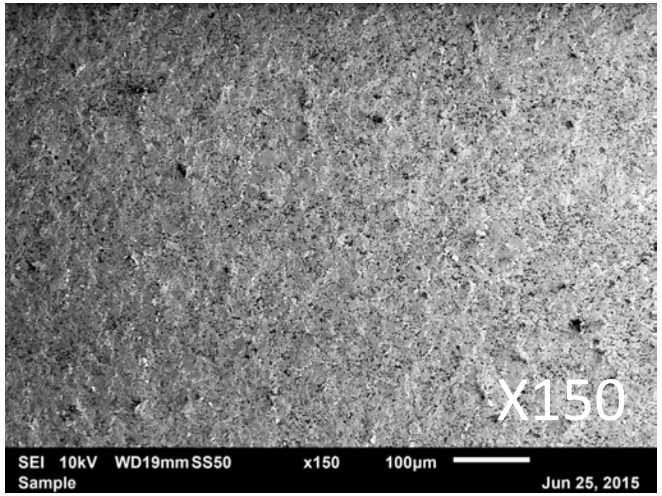
Stretch Cycles versus Stretch Percentage

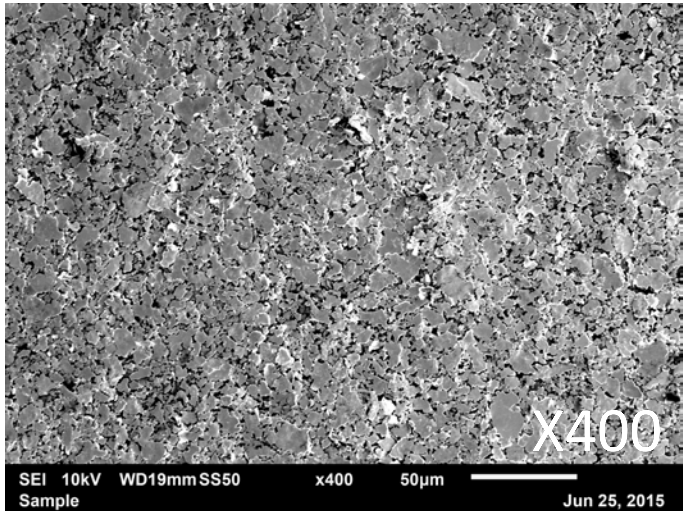


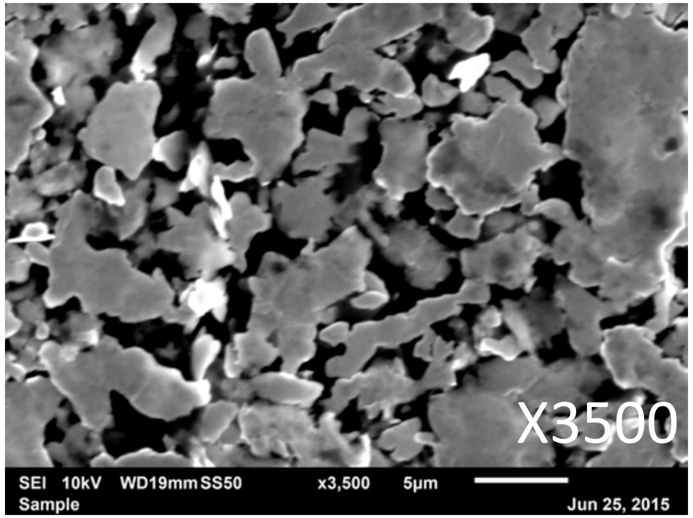
*Failure criterion based on 1000 ohm maximum resistance or initiation of resistance spike

Analysis of the Printed Ink at 50% Stretch

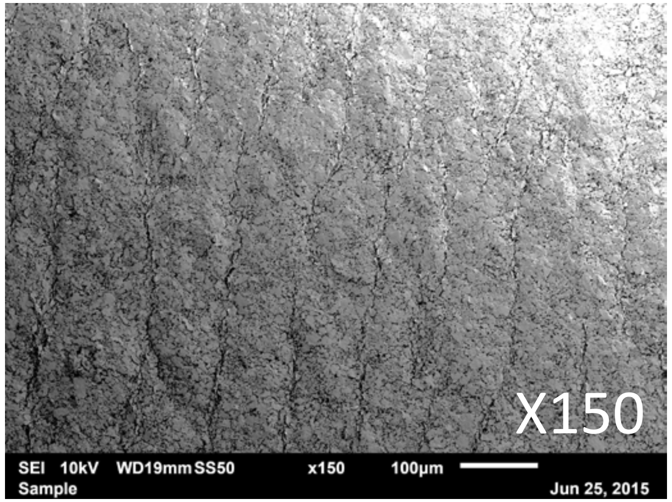
Film A 5 mil

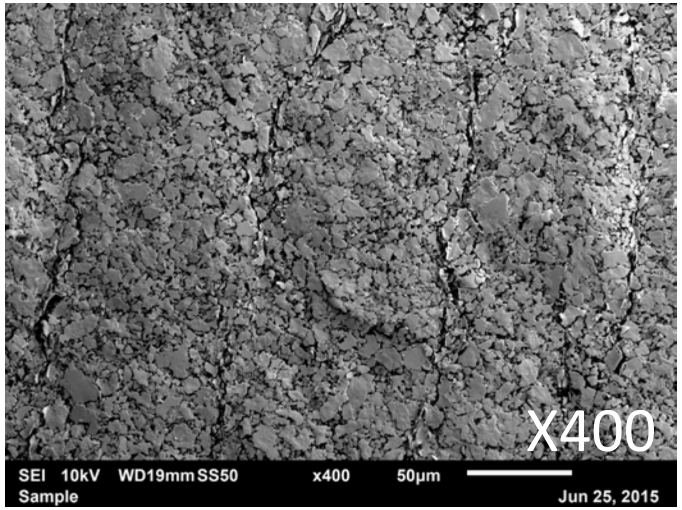


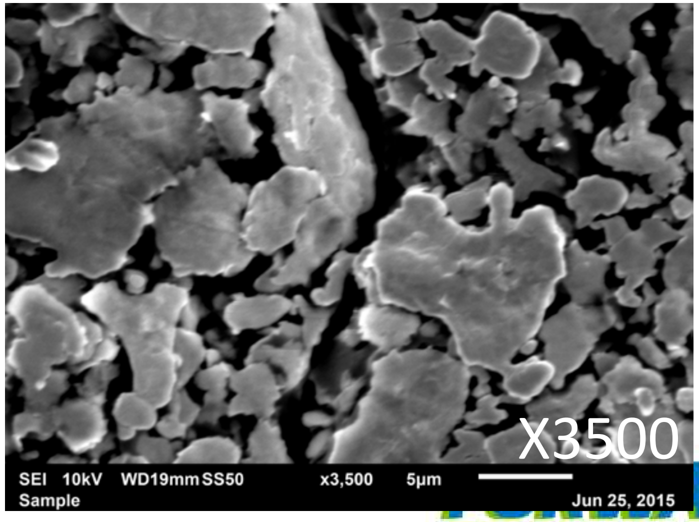




Film B 4 mil

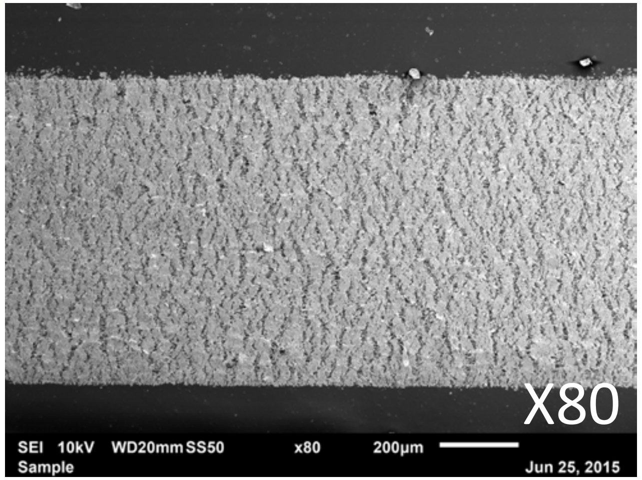


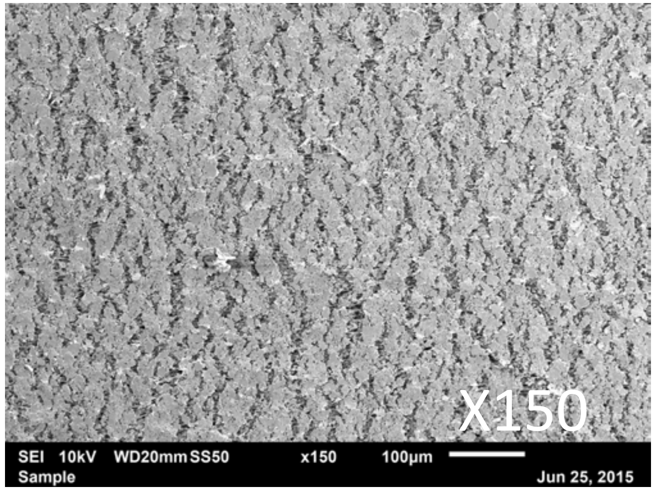


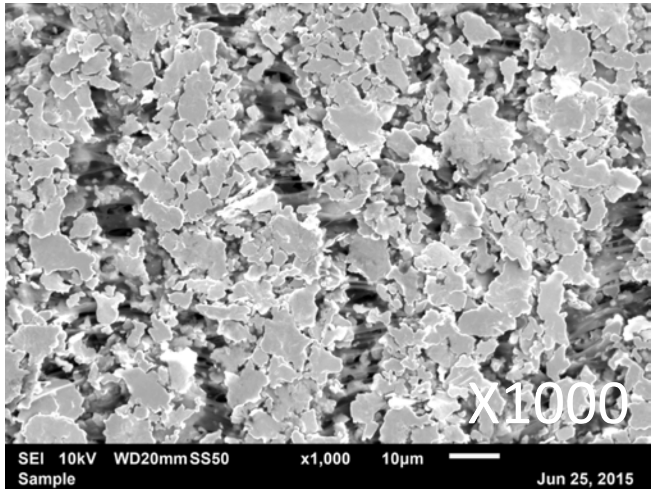


Analysis of the Printed Ink at 100% Stretch

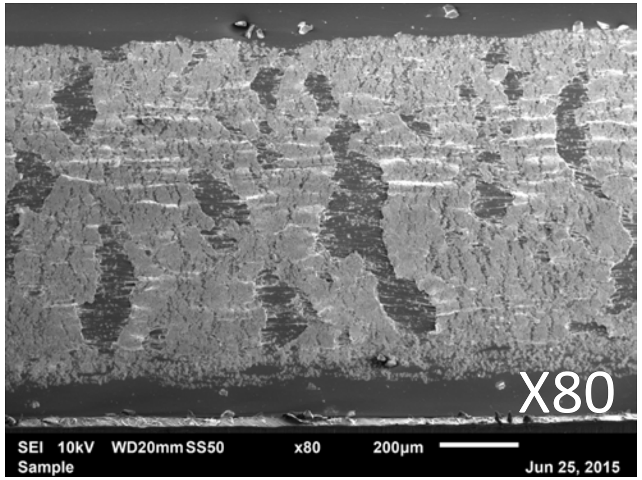
Film A 5 mil

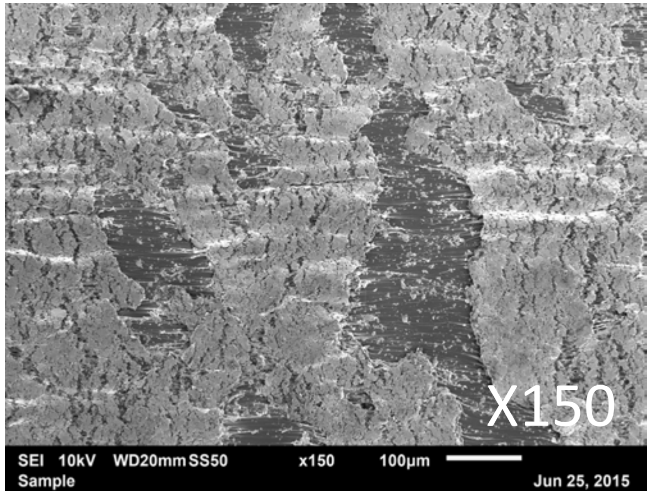


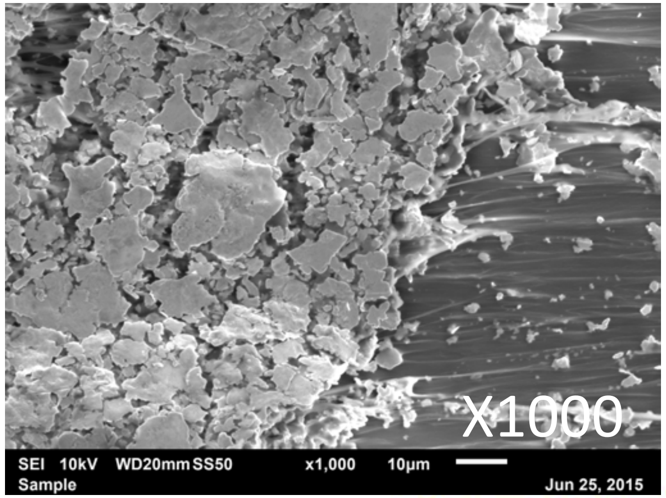




Film B 4 mil

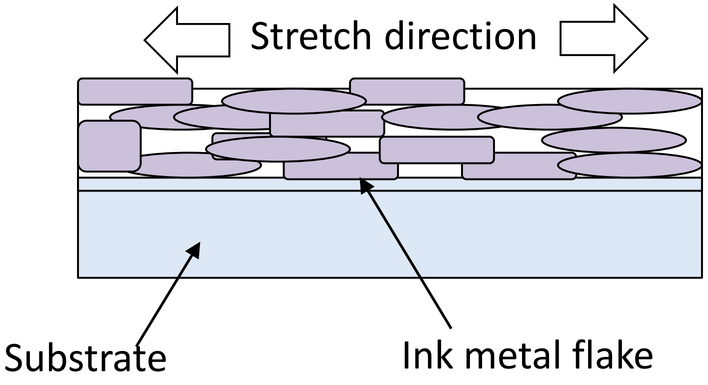






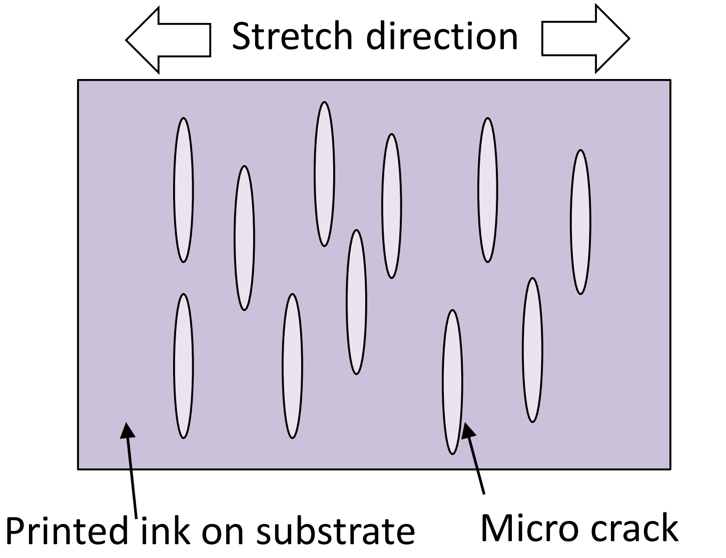
Print Ink Stretch Mechanisms

Low stretch strain



Metal fakes slide against each other to accommodate the strain during stretch

High stretch strain



Microcracks are formed as a strain relieve during stretch

- Film A: Strong bonding between film and printed ink. More rigid substrate may be able to hold the metal flakes in place during stretch and stress can be relieved through particle sliding or microcrak formation to maintain conductivity
- Film B: weak bonding between film and printed ink. More rigid substrate may cause more debonding, therefore sharper increase in resistance

Summary and Conclusions

- Stretchability of a conductive ink is evaluated to understand its behavior and performance at different stretch strains
- Test coupon is designed and validated to facilitate the stretchability testing and eliminate resistance measurement instability during test
- Different designs, materials and test parameters affecting ink stretchability are evaluated:
    - Design pattern
    - Ink line width
    - Stretch speed
    - Stretch percentage
    - Substrate material and thickness
- The ink shows a continued increase in electrical resistance with stretch strains and with stretch cycles. Resistance spikes tend to occur after prolonged stretch cycles
- Higher stretch speed (strain rate) causes more instability of printed ink resistance during stretching
- Substrate material and thickness greatly affect ink stretchability. More fundamental studies are needed to understand the effect
- The stretchability mechanisms are explained in terms of sliding between metal flakes and formation of microcracks within the printed ink matrix
- Based on the experimental data, the expected life cycle versus stretch strain is plotted for the ink studied

Future Work

- Evaluate the effect of following design and test factors on stretchability
  - Dwell time
  - Ink thickness
  - Carbon overcoat
  - Encapsulation
- Obtain fundamental understanding of substrate effect on stretchability to assist substrate selection (Rigidity, bonding strength)
- Perform DoE experiments on stretchability and understand interactions between different factors
- Standardize stretchability test methods and recommend test coupon design, through IPC D65 standard subcommittee

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Thank You for Your Attention!