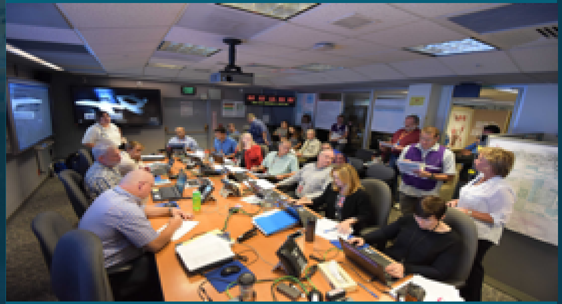
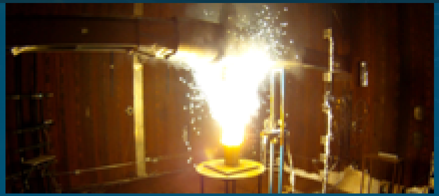
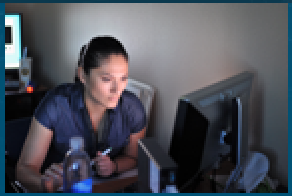
SAND2022-12788C

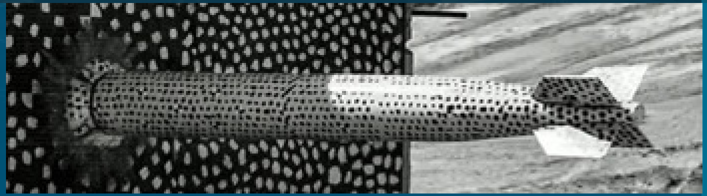
Sandia National Laboratories

Late E-2504 Electrodeposition of Re on Aerosol Jet Printed Metal Seed Layers on Flexible Substrates









Presented By

Lok-kun Tsui,[1-2] Kamal Ahammed,[3] Yongkun Sui,[2] Emily A. Weigel,[2] Qiang Huang,[3] and Judith M. Lavin[2]

[1] University of New Mexico, Center for Micro-Engineered Materials, Albuquerque, NM, USA

[2] Sandia National Laboratories, Albuquerque, NM, USA

[3] University of Alabama, Chemical and Biological Engineering, Tuscaloosa, AL, USA

CENTER FOR MICRO-ENGINEERED MATERIALS

THE UNIVERSITY OF ALABAMA

ATLANTA GEORGIA

242nd ECS Meeting

October 9-13, 2022

Atlanta Hilton

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

SANDXXXXXX

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Presentation Overview

I. Introduction
- Motivation: Superconducting Connections for Quantum Computers.
- Aerosol Jet Printing (AJP) as a patterning method for electrodeposition of Rhenium

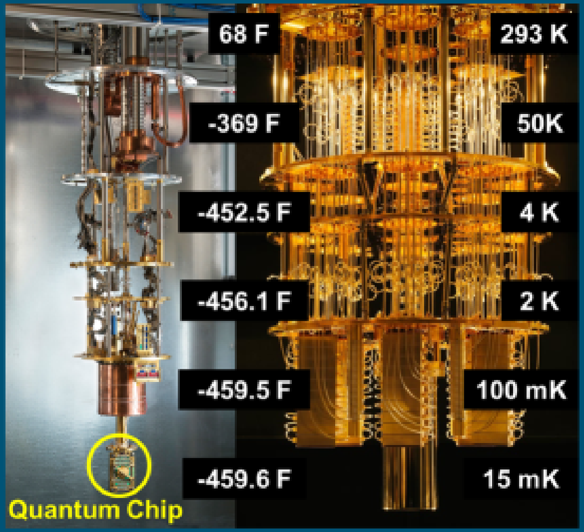
II. Results and Discussion
- AJP of Ag and Au on Kapton
- Film Adhesion
- Superconducting Behavior
- Flexure Testing

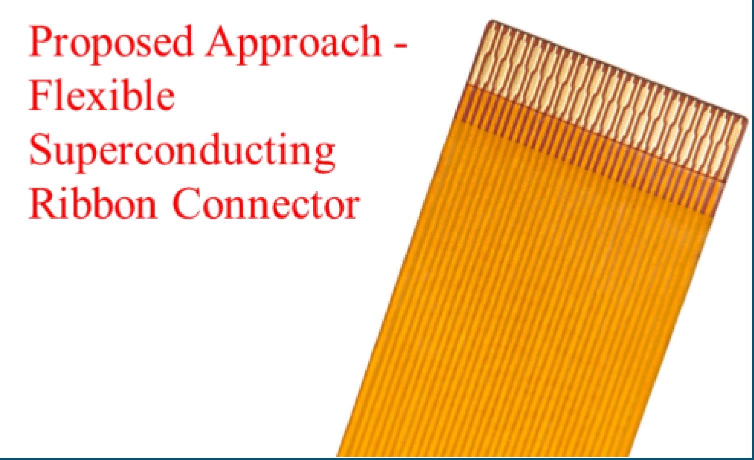
III. Conclusions and Future Work

Motivation: Additive Manufacturing for Coreless Flyback Transformers

- **Quantum Computers** (QCs) can process algorithms to solve currently intractable problems in physics, chemistry, cryptography, medicine, and finance. [1]
- Quantum chips require cooling by dilution refrigeration to 10 mK.
- Interconnection to the quantum chip are typically implemented using a chandelier of stainless steel cables that must be individually and manually assembled.
- A ribbon of printed superconducting material with a critical temperature above 4 K eliminates Joule heating in liquid He cooled parts of the chandelier improves the thermal efficiency.
- Flexibility also enables more compact and easier to build designs.
- **Project Objectives**: Develop a process to pattern superconducting material on flexible substrates.

Quantum Computer Chandelier

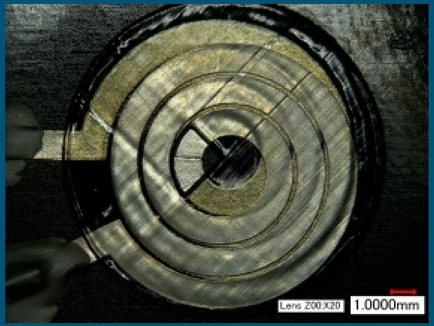




[1] F. Bova, A. Goldfarb, R.G. Melko "Commercial Applications of Quantum Computing." EPJ Quantum Tech. (2021) 8, 2.

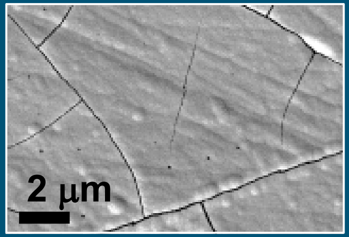
Previous Work

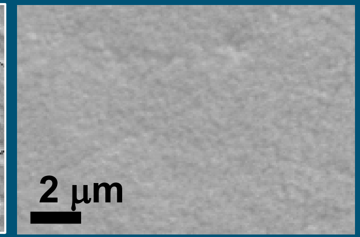
Sandia National Labs:
Aerosol Jet Printing + Electrodeposition [1]



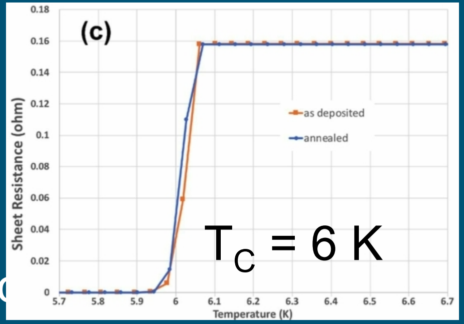
- Aerosol Jet Printing is capable of printing metal and insulator features at < 10 μm resolution on a wide range of rigid and flexible substrates.
- AJP of Ag as a seed layer was demonstrated with electrodeposition Cu and Ni for improved electrical conductivity.

University of Alabama:
Water-in-Salt Method [2]





Aqueous W-I-S Metho



- The Water-in-Salt method for electrodeposition of Re suppresses hydrogen evolution and improves film quality while traditional aqueous electrolytes for Re deposition produced cracked films.[2]
- Amorphous Re on Au substrates had a TC of 6K.

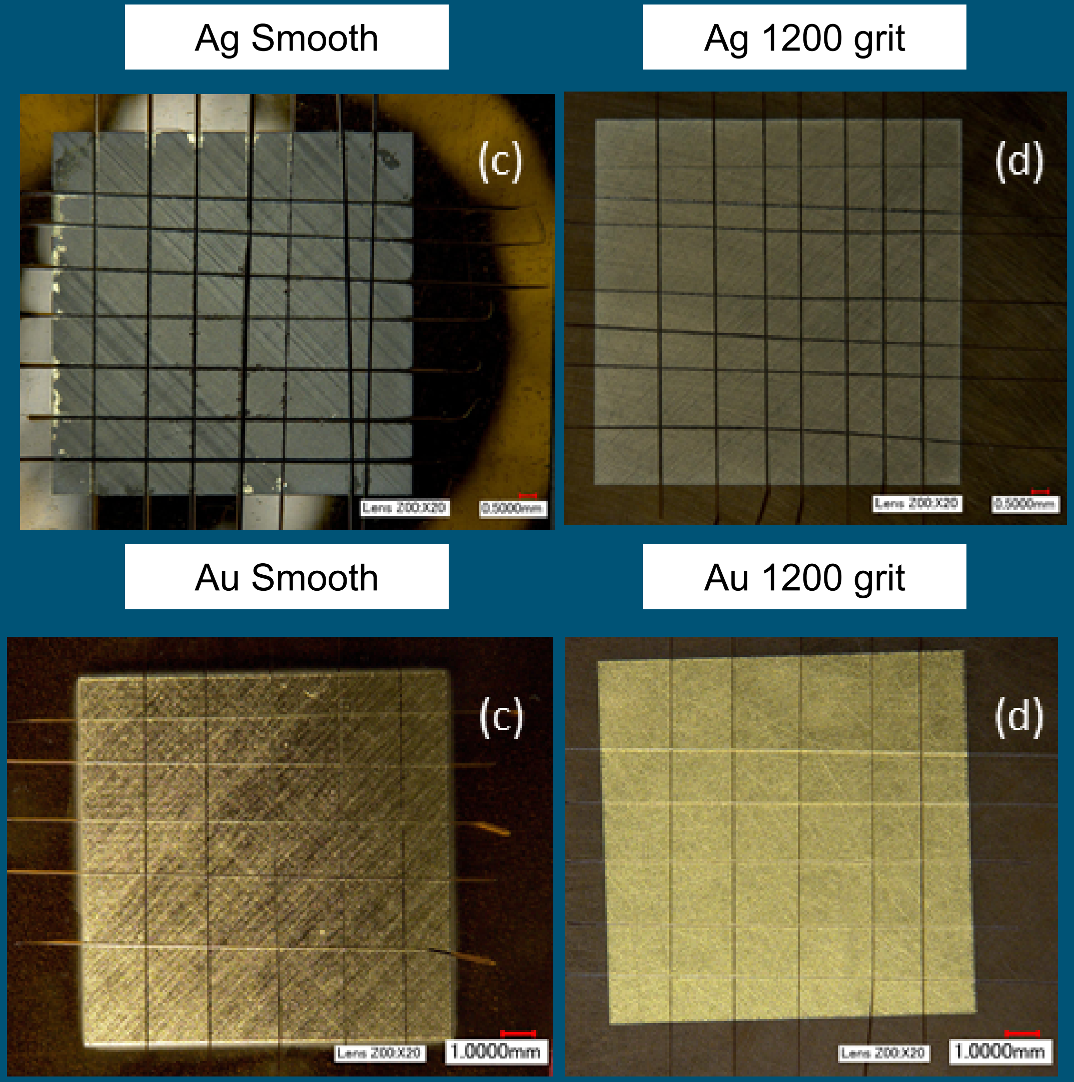
See also: G01-1083 – "Chemistry Effects on the Electrodeposition of Re, Co, and Alloys" presented by Q. Huang.

A combination of both approaches should enable the patterning of superconducting Re on flexible substrates.

[1] L. Tsui, S. C. Kayser, S. A. Strong, J. M. Lavin ECS J. Solid State Sci. Technol., 2021, 10, 047001
[2] Q. Huang & Y. Hu J. Electrochem. Soc., 2018 165, D796

Adhesion Studies of Au and Ag on Kapton

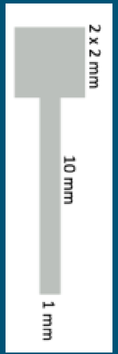
- Aerosol Jet Printing of UTDots Ag and Au nanoparticle inks was performed on 127 µm thick Kapton substrates.
- Ag and Au were sintered in air at 120°C and 300°C respectively.
- Substrates were tested as received and roughened with 1200 grit sandpaper to improve adhesion.
- ASTM D3359 tape test showed Ag on roughened substrates had superior (5B rating, 0% loss) adhesion compared with Ag on smooth substrate (4B rating, 5% loss).
- Au on both smooth and roughened substrates had equally good adhesion (5B rating, 0% loss).



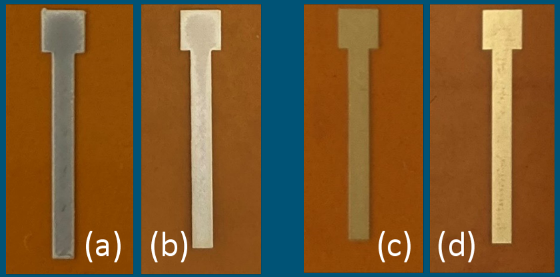
Seed Layer Preparation

- 1 mm x 10 mm strips with a 2 x 2 mm contact pad were printed with Au and Ag inks.

Design | Ag | Au



Smooth 1200 gr. Smooth 1200 gr.



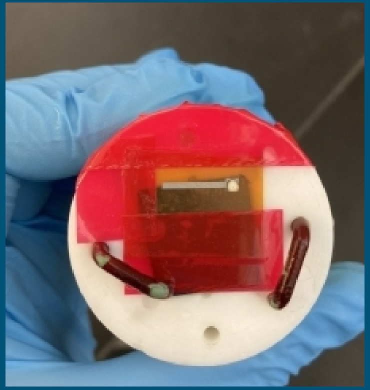
- Seed Layer Thickness:

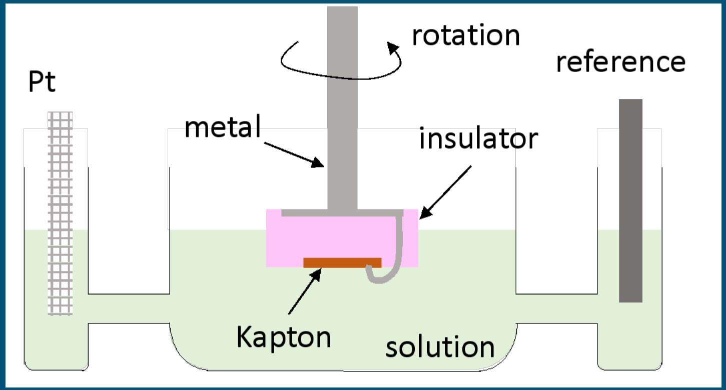
	Smooth	1200 grit
Ag	4.16 µm	3.26 µm
Au	0.85 µm	1.62 µm

- 5 x 5 mm pads were also prepared for adhesion studies.

Electrodeposition of Re

- Electrodeposition of Re was carried out in: 25 mM NH_4ReO_4, 0.1 M H_2SO_4, 5 M LiCl in a RDE setup at 400 RPM.
- Reference Electrode: Ag/AgCl
- Counter Electrode: Pt

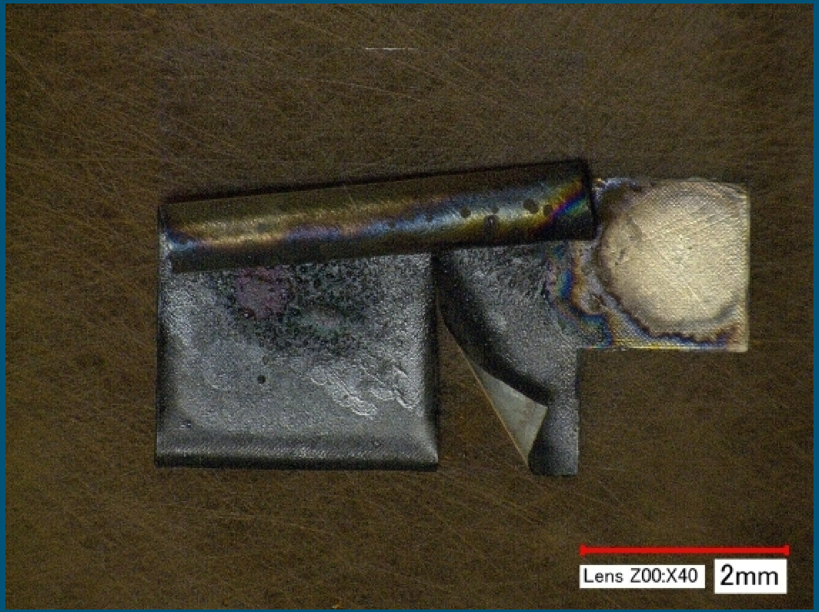




Electrodeposition of Re on Ag

- Deposition of Re on Ag was studied first.
- DC deposition resulted in poor adhesion of resulted in complete delamination of both the Ag seed layer and electrodeposited Re.
- Deposition on 1200 grit roughened Kapton marginally improved adhesion, but samples still delaminated.
- Adhesion test structure showed partial delamination prior to grid cutting. Assigned it a 0B rating (> 65% material loss).
- While Ag on Kapton alone showed good adhesion, stresses applied during electrodeposition easily caused the films to peel off.
- Pulse deposition (-1V for 30 seconds, OCV for 30 seconds) was also unsuccessful in mitigating delamination.

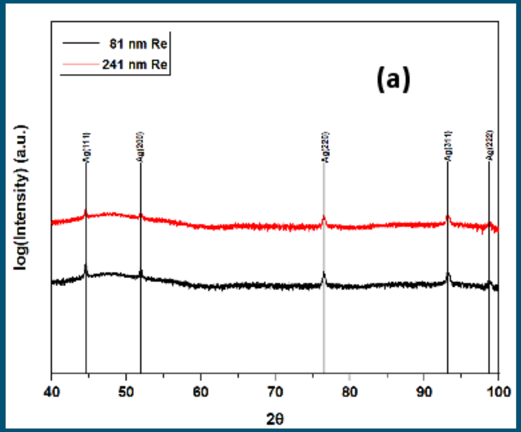
Delamination of Re + Ag strip device

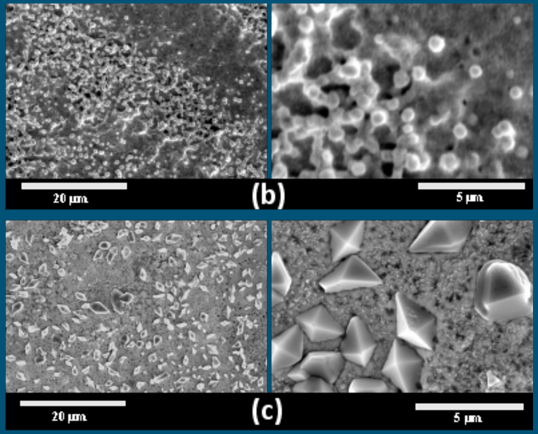


Partial delamination of Re + Ag on adhesion test part (1200 grit Kapton).

Film Characterization (Re + Ag)

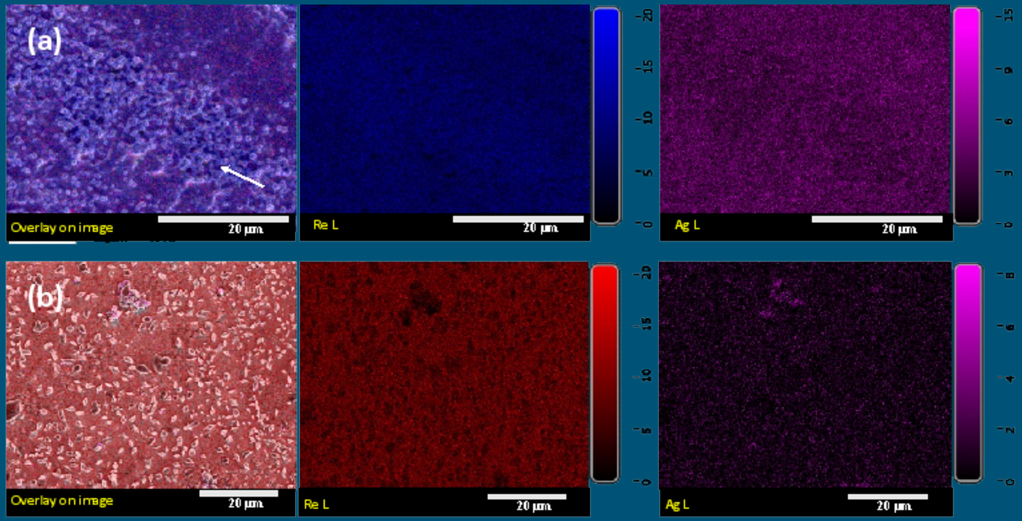
- X-ray diffraction of electrodeposited Re films on Ag substrates show that the films are amorphous. Only peaks corresponding to Ag are observed.
- Amorphous phase is desirable because of the higher critical temperature for superconductivity.
- SEM images show that Re deposits in the form of nodules on the surface of Ag.
- SEM EDS measurements complete coverage of the sample with Re.
- The nodular structure suggests difficulty for Re on Ag to nucleate and grow into a uniform film.





81 nm Re + Ag

241 nm Re + Ag



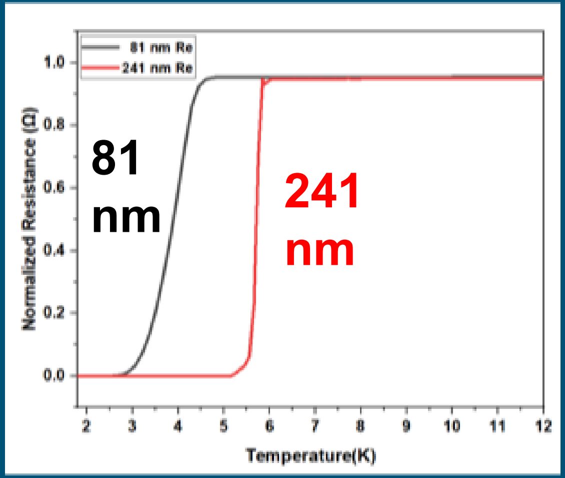
81 nm Re + Ag

241 nm Re + Ag

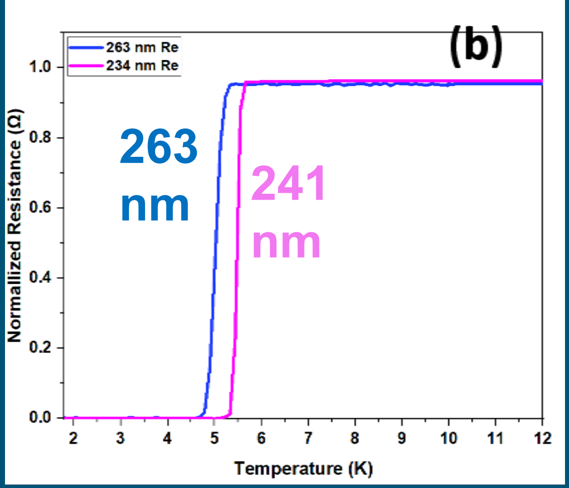
Electrical Characteristics & Superconductivity (Re + Ag)

- The critical temperature was measured using a Physical Properties Measurement System (PPMS) from 30 K to 1.8 K.
- Samples needed to be taped down to overcome delamination.
- Thick layer of Re shows a sharp transition to superconductivity at 5.5 K. Thin layers of Ag show a broader superconducting transition curve across 3-5 K.
- May be due to higher local variability in film thickness of the thin samples.
- Pulse electrodeposition of Re also resulted in samples with a sharp transition temperature between 4.5-5.5 K.
- While these results confirm successful formation of superconducting Re by electrodeposition on AJP seed layers, poor adhesion makes Ag an impractical material for use.

DC Electrodeposited Re on Ag



Pulse Electrodeposition of Re on Ag

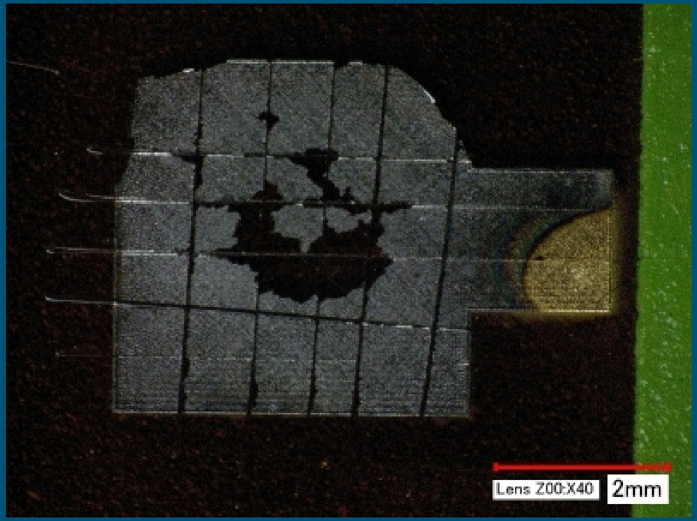


Electrodeposition of Re on Au

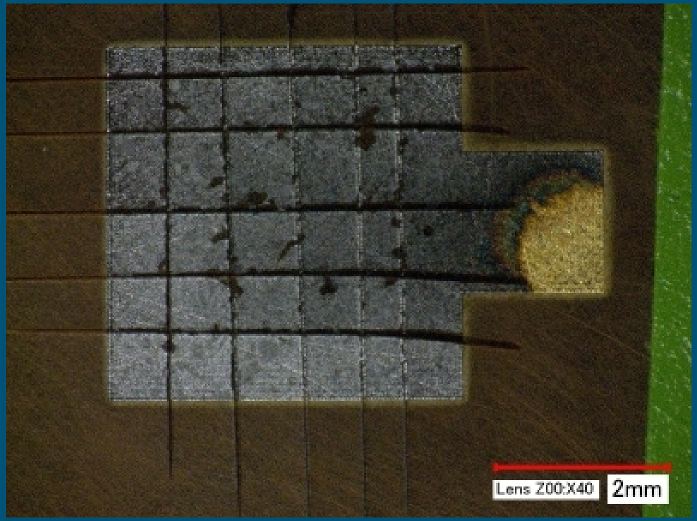
- Deposition of Re on Au was studied next.
- DC plating at -1V for 300 seconds was performed and a Re film thickness of ~400 nm was targeted.
- Re on Au strip structures on both smooth and 1200 grit roughened Kapton substrates showed no delamination after plating.
- Adhesion test structures were successfully measured with the ASTM D3359 Tape Test:
  - Smooth Kapton: 2B (15-20% loss)
  - 1200 Grit Kapton: 4B (5% loss)
- Increased cost of Au vs. Ag ink would be a tradeoff for improved adhesion.

Re + Au
Strip device

Re + Au (Smooth Kapton)

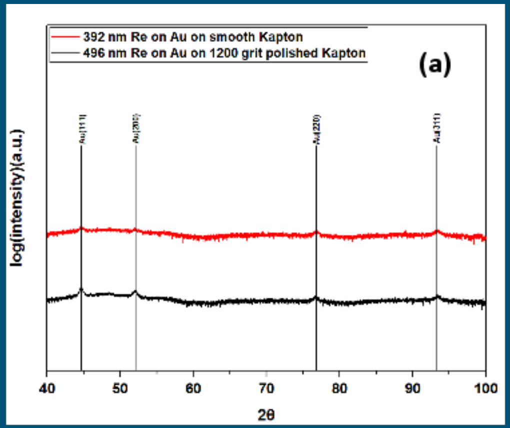


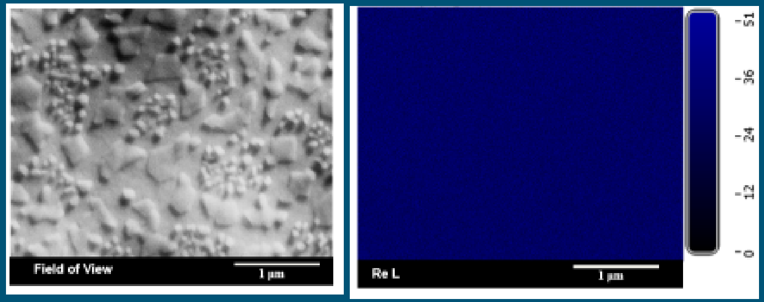
Re + Au (1200 gr. Kapton)

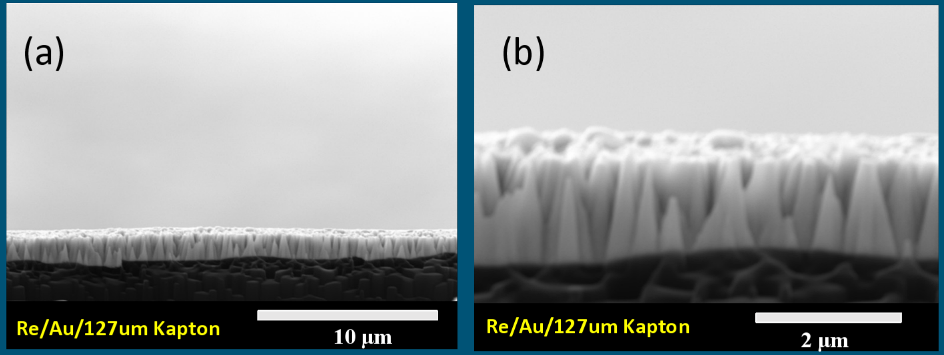


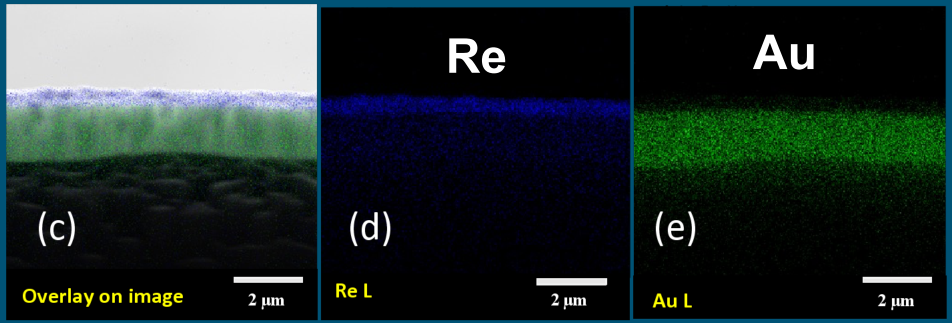
Film Characterization (Re + Au)

- XRD of 400-500 nm of Re on Au shows that the film is amorphous for both smooth and 1200 grit Kapton substrates. Only peaks detected are from Au seed layer.
- Top-view SEM shows that Re forms as islands on the surface of the Au seed layer.
- Top-view EDS shows that the entire surface is covered by Re as a continuous film.
- Cross sectional SEM and EDS shows approximately 1 micron of Au in columnar structure with a cohesive layer of Re on top.







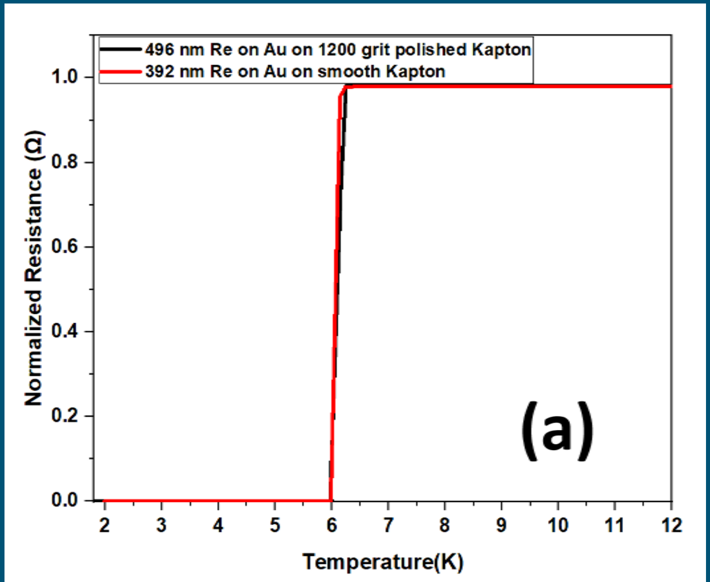


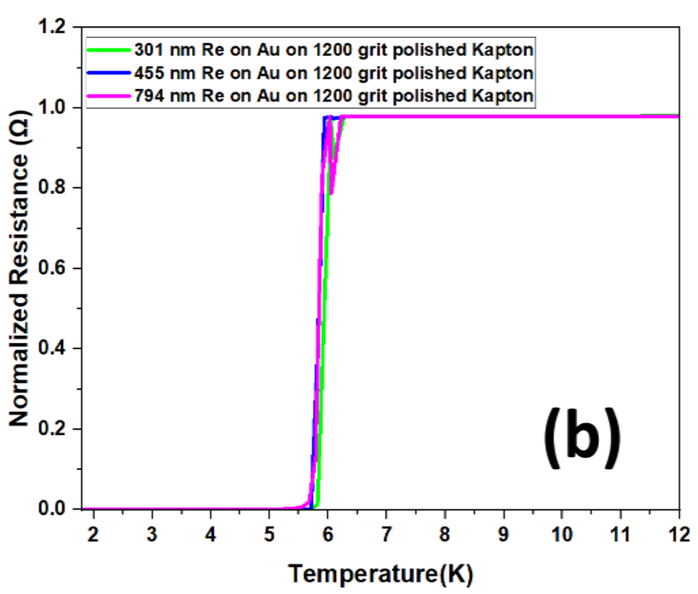
Electrical Characteristics & Superconductivity (Re + Au)

- Addition of Re to Au slightly decreases the room temperature resistance of the strip devices at 500 nm.

Substrate	Re Thickness from XRF (nm)	Resistance (Ω) Before Re Deposition	Resistance (Ω) After Re Deposition
Au on 1200 grit rough Kapton	496	0.169	0.164
Au on smooth Kapton	466	0.158	0.154

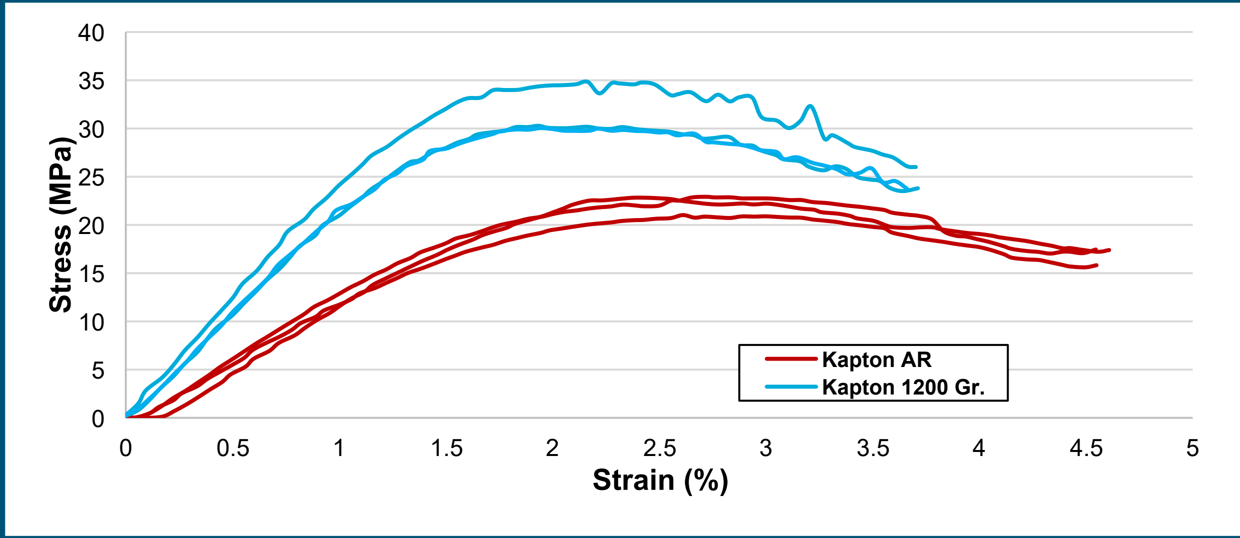
- Deposition on smooth Kapton and 1200 grit rough Kapton resulted in a superconducting T_C of 6 K.
- Film thicknesses of 201, 455, and 794 nm of Re on 1200 grit rough Kapton showed a consistent superconducting TC of 6K.
- These results show the T_C is better controlled and reproducible on Au substrates vs. Ag

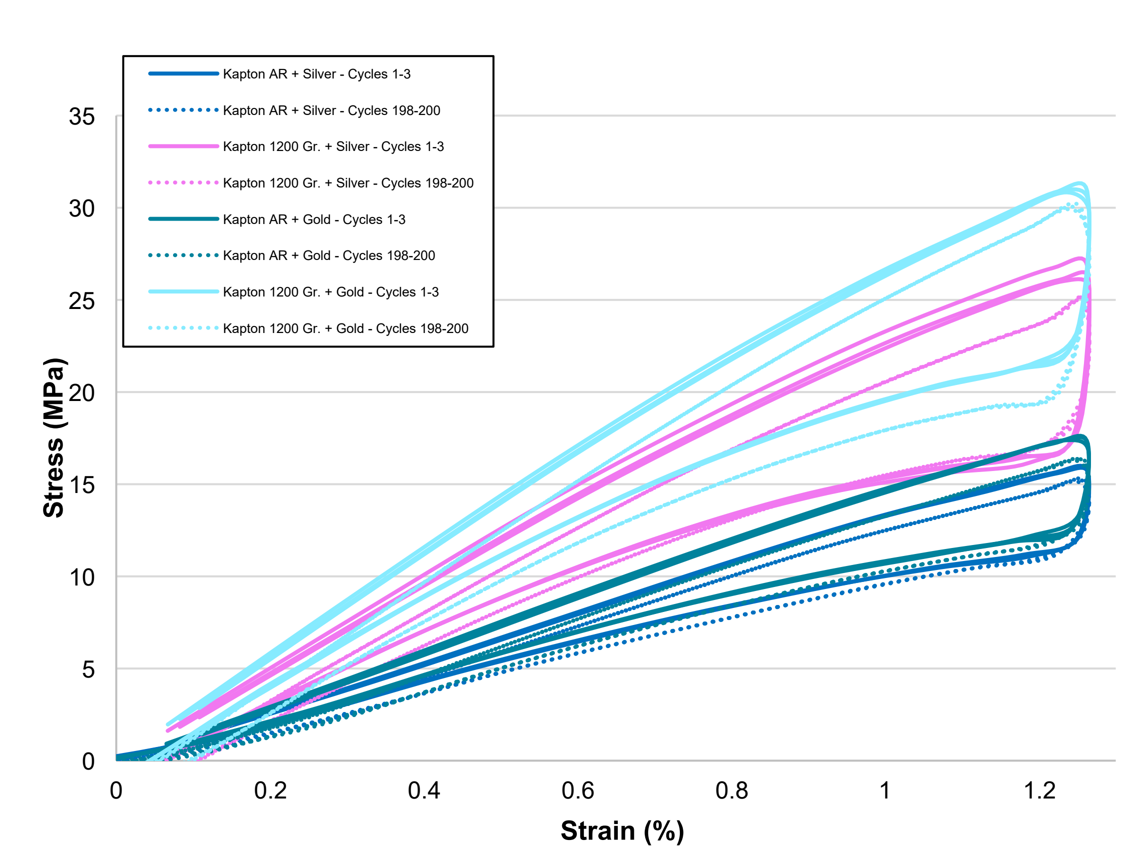




Flexure Testing

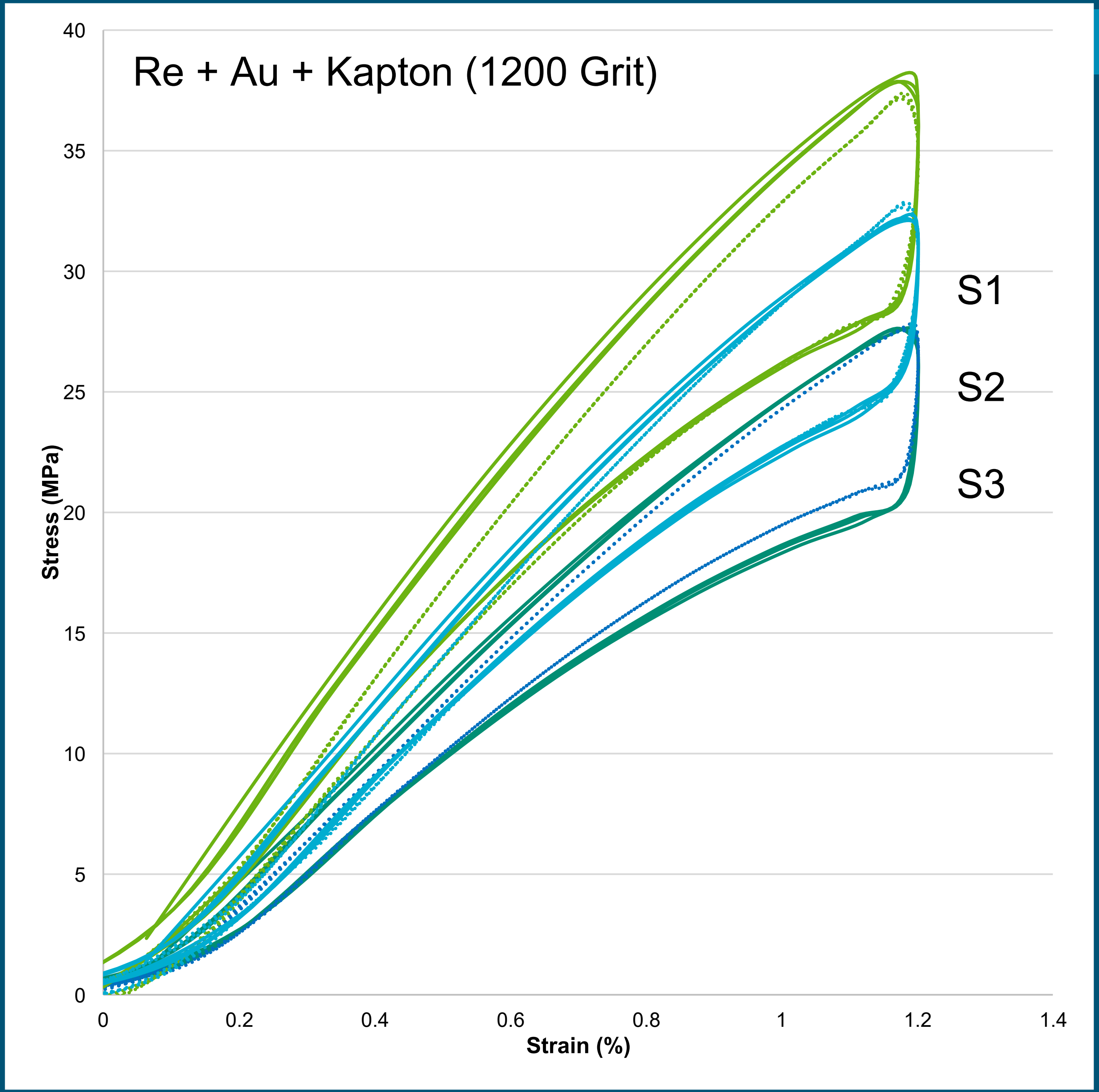
- Three point bend testing was performed on the substrates, with seed layers, and with electrodeposited Re. Cycling testing was also performed for 200 cycles.
- Kapton after roughening to 1200 grit reaches a higher stress for the same strain.
- The films which are unpolished reach a lower stress of 15-17 MPa at 1.25% strain while the polished samples have around 27-30 MPa.
- Seed layers are < 5 µm vs. 127 µm thick substrates-- The flexural properties of the substrates dominate over the presence of the seed layer.
- A less than 1 MPa shift downward in stress at 1.25% strain is observed between the first 3 cycles and last 3 cycles.
- Flexure testing of Re + Au + on 1200 grit roughened Kapton substrate showed at most a 5 MPa increase in stress at 1% strain.





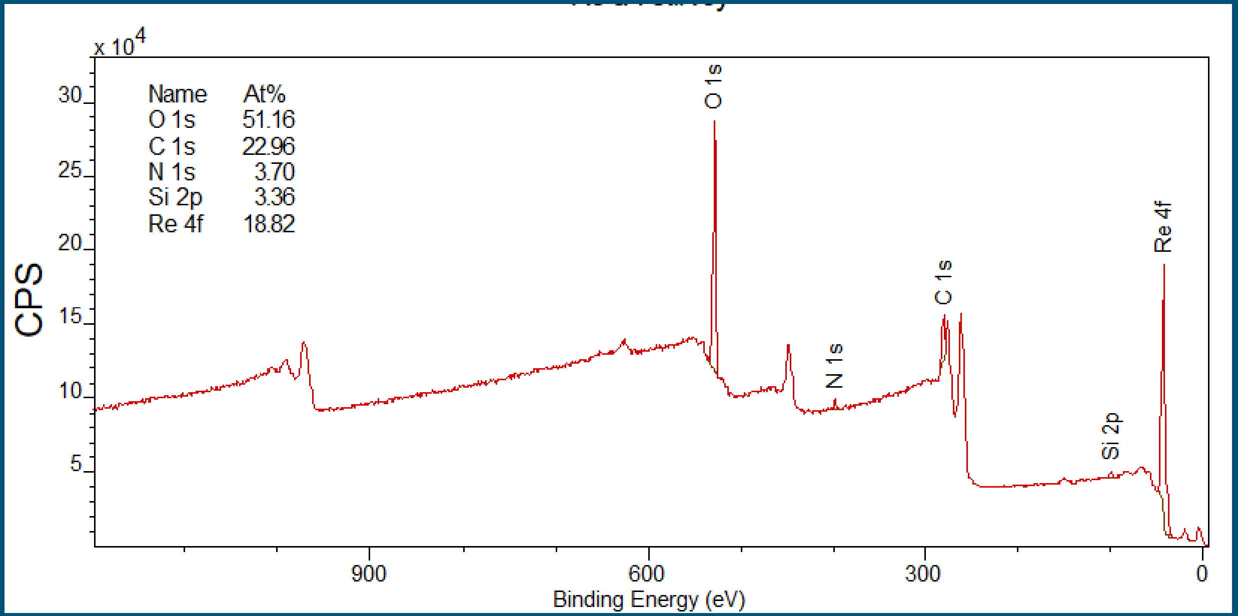
Flexure Testing

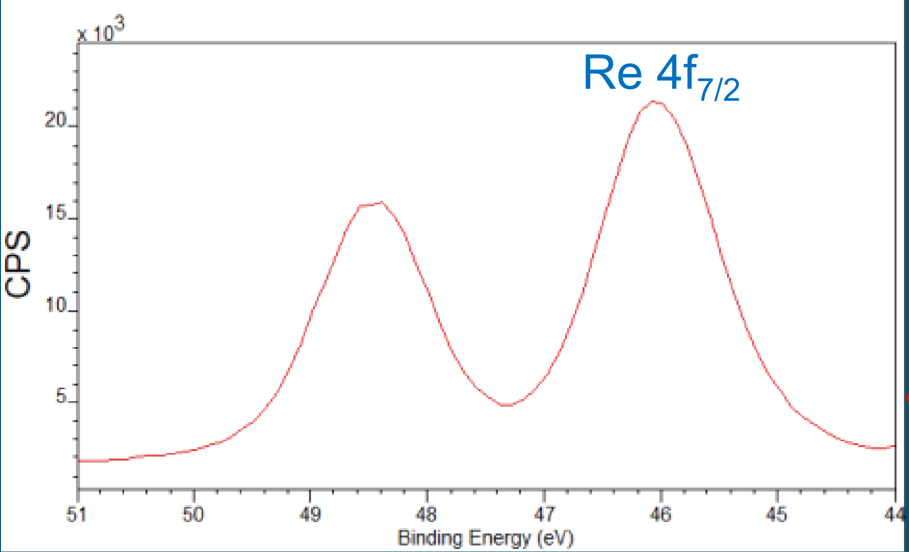
- Three point bend testing was performed on the substrates, with seed layers, and with electrodeposited Re. Cycling testing was also performed for 200 cycles.
- Kapton after roughening to 1200 grit reaches a higher stress for the same strain.
- The films which are unpolished reach a lower stress of 15-17 MPa at 1.25% strain while the polished samples have around 27-30 MPa.
- Seed layers are < 5 μm vs. 127 μm thick substrates-- The flexural properties of the substrates dominate over the presence of the seed layer.
- A less than 1 MPa shift downward in stress at 1.25% strain is observed between the first 3 cycles and last 3 cycles.
- Flexure testing of Re + Au + on 1200 grit roughened Kapton substrate showed at most a 5 MPa increase in stress at 1% strain.



Surface Oxidation of Re During Ambient Air Storage

- During transfer of samples between SNL and UA, darkening of the Re films were observed.
- Effect was more prominent during humid monsoon season.
- XPS measurements with penetration depth of 10 nm indicates oxidation of Re(0) to ReO_3.
- Further transfers of Re between labs required packaging in inert (N_2, < 1 ppm O_2 and H_2O) and vacuum atmosphere bags.
- For practical QC applications, encapsulation of Re films in a protective layer will be required.





$4f_{7/2}$ Binding Energies for Oxidation States of Re

Species	B.E. (eV)	Std. Dev.
Re(0)	40.5	0.3
ReO2	43.0	0.6
ReO3	45.4	1.3
Re2O7	46.7	0.1
KReO4	46.1	
K2[ReCl6]	44.1	0.2

Conclusions

- We have demonstrated the capability of patterning flexible superconducting Re for QC interconnects using a combined AJP / Electrodeposition process.
- Au seed layers exhibited superior adhesion to flexible Kapton substrate after electrodeposition of Re.
- Substrate independent and thickness independent superconducting T_C of 6 K was measured. T_C preserved after flexure testing.
- Oxidation in ambient atmosphere remains an unsolved challenge.

Future Work:

- Develop methods to protect against oxidation by encapsulation.
- Characterize and optimize the high frequency response of printed Re superconductors.

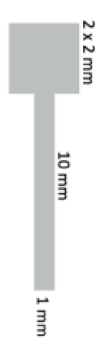
Acknowledgements

We acknowledge Shianne Carroll (Sandia National Labs) for performing the three point bend flexure tests and Angelica Benavidez (UNM CMEM) for performing the XPS measurements.

Funding:

This work was funded by Sandia National Laboratories. *Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.* **SANDXXXXX**

Digital Design



Aerosol Jet Printing + Electrodeposition

Au + Kapton

Re

T_C > 6 K Superconductor