Passive Wireless Sensors Fabricated by Direct-Writing for Temperature and Health Monitoring of Energy Systems in Harsh-Environments

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2021 Crosscutting Research and Advanced Energy Systems Project Review Meeting
SENSORS & CONTROLS PROGRAM
May 19, 2021 (8:30 am)

Award DE-FE0026171

Initial Acknowledgments:

Acknowledgment: "This material is based upon work supported by the Department of Energy Award Number DE-FE0026171"

Background- Harsh Environment Sensing Needs

In-situ monitoring of high temperature systems in extreme conditions encountered in: Chemical Synthesis; Ceramic Processing; Metallurgy/Manufacturing Industries, etc.

Harsh Environments

- High temperature (>500ºC).
- High pressure (up to 1000 psi).
- Various pO_2 levels.
- Corrosive conditions (molten inorganics or reactive gasses).

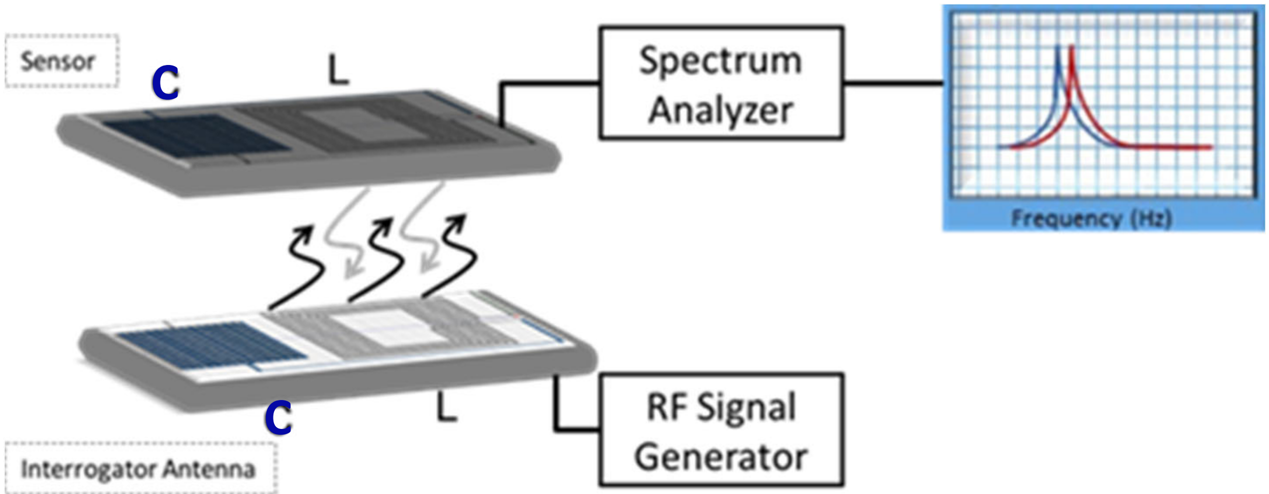
Ability to monitor:

- ✔ Temperature.
- ✔ Stress/strain components.
- ✔ *In situ* failure analysis.

Current Issues

1) Stability (Material property), 2) Sensor integration with the sensing system (Interface), 3) Reading sensor response (interconnection)

Sensing Mechanism (LC-based RF Resonator)



Sensor Components:
Planar **L (inductor)** and **C (capacitor)** deposited on a dielectric substrate.

Working Mechanism:

$\Delta T \ \alpha \ \Delta f_r$

$$f_r = \frac{1}{2\pi\sqrt{LC}} \Rightarrow C = \frac{\varepsilon_0 \varepsilon_r A}{d}$$

ε_0 – Constant
A; d – Predefined variables
ε_r – Relative Permittivity
(ε_r α T)

Processing Vision – Peel and Stick Sensor

Item A represents the organic carrier film.

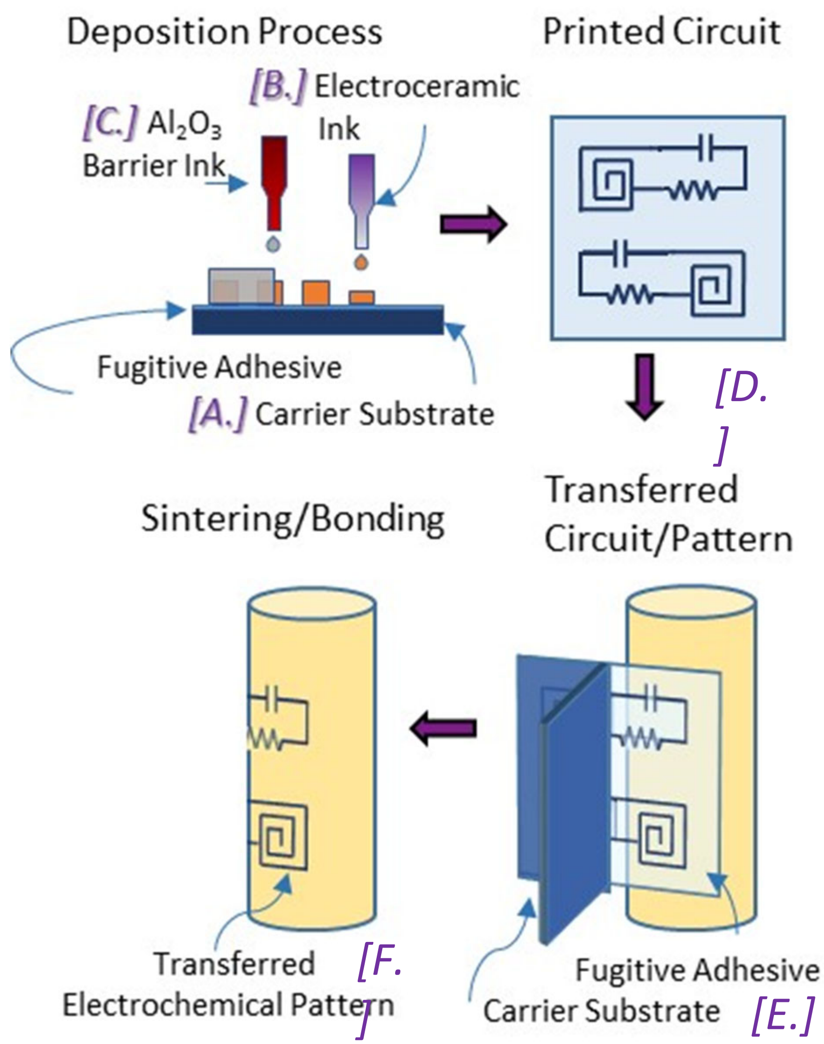
Item B represents the polymer-precursor ink (converts to an electroceramic after heat treatment).

Item C represents a possible barrier layer.

Item D represents RF circuit sensor circuit printed on the transfer paper.

Item E shows the RF circuit pattern being placed upon the energy-system component.

Item F represents the pyrolysis of the organic carrier and bonding.



Program Objectives

Task 2:

- Investigate phase formation, sintering/grain growth, and electrical properties of polymer-derived ceramic (PDC) composites between 500-1700 °C.

Task 3:

- Define processes to fabricate sensor through direct-writing (or micro-casting) electroceramic composites.
- Develop methods to form monolithic "peel-and-stick" technology.

Task 4:

- Design of RF passive wireless LCR circuits and receiver (reader) antennas for testing at temperature up to 1700°C.

Task 5:

- Demonstrate the passive wireless sensor system on a SOFC repeat unit and a singular gas turbine blade prototype as example applications *(currently delayed due to sensor limitations; new sensor designs required).*

Task 2.0: Fabrication and Characterization of Polymer-Derived Electroceramic Composites. (Sabolsky, Varadharajan)

Task 2.0 Objective:

Investigate phase formation, sintering/grain growth, and electrical properties of electroceramic composites for applications at 500-1500 °C.

- **System 1:** Indium tin oxide (ITO)
- **System 2:** Lanthanum strontium chromate (LSC)
- **System 3:** Polymer Derived Ceramics (PDCs)

Task 2.0 Approach:

- **Subtask 2.1 Analysis of Multifunctional Electroceramic Composites:**

Electrically conducting/semi-conducting oxides namely, indium tin oxide (ITO), and lanthanum strontium chromate (LSC) will be investigated as precursors to fabricate various electroceramic compositions. This will include the addition of fillers to understand the stability.

- **Subtask 2.2 Thermal Processing of Composite Compositions:**

Samples will be pressed into bars, and also screen-printed onto Al_2O_3 or ZrO_2 dense substrates (for initial observation on shrinkage effects). Thermal processing the materials in various atmospheres (air, inert, reducing atmosphere) will be completed up to 1700^oC.

- **Subtask 2.3 Composite Material Testing and Characterization**

Electrical testing (at high-temperature) and phase/chemistry characterization will be completed on the polymer-derived electroceramic composites.

Task 2.0 Current status:

- **Subtask 2.1 Synthesis of Multifunctional Electroceramic Composites:**
  - ✔ ***System 1:*** Indium tin oxide (ITO).
  - ✔ ***System 2:*** Lanthanum strontium chromate (LSC).
  - ✔ ***System 3:*** Polymer Derived Ceramics (PDCs).

- **Subtask 2.2 Thermal Processing of Composite Compositions:**
  - ✔ Investigated the thermal processing of the oxide systems on Al_2O_3 substrates.

- **Subtask 2.3 Composite Material Testing and Characterization:**
  - ✔ Phase/chemistry characterization is completed on the polymer-derived and oxide electroceramic composites.

Electroceramic Material Systems for LC Resonator

Material systems were evaluated based on:

- Operating temperature (500 – 1000°C; 1000 – 1700°C)
- Electrical Conductivity (high conductivity; positive temperature coefficient)
- Thermal Stability (No/minimal change in microstructure)
- Compatibility with the substrate (no interfacial reaction)

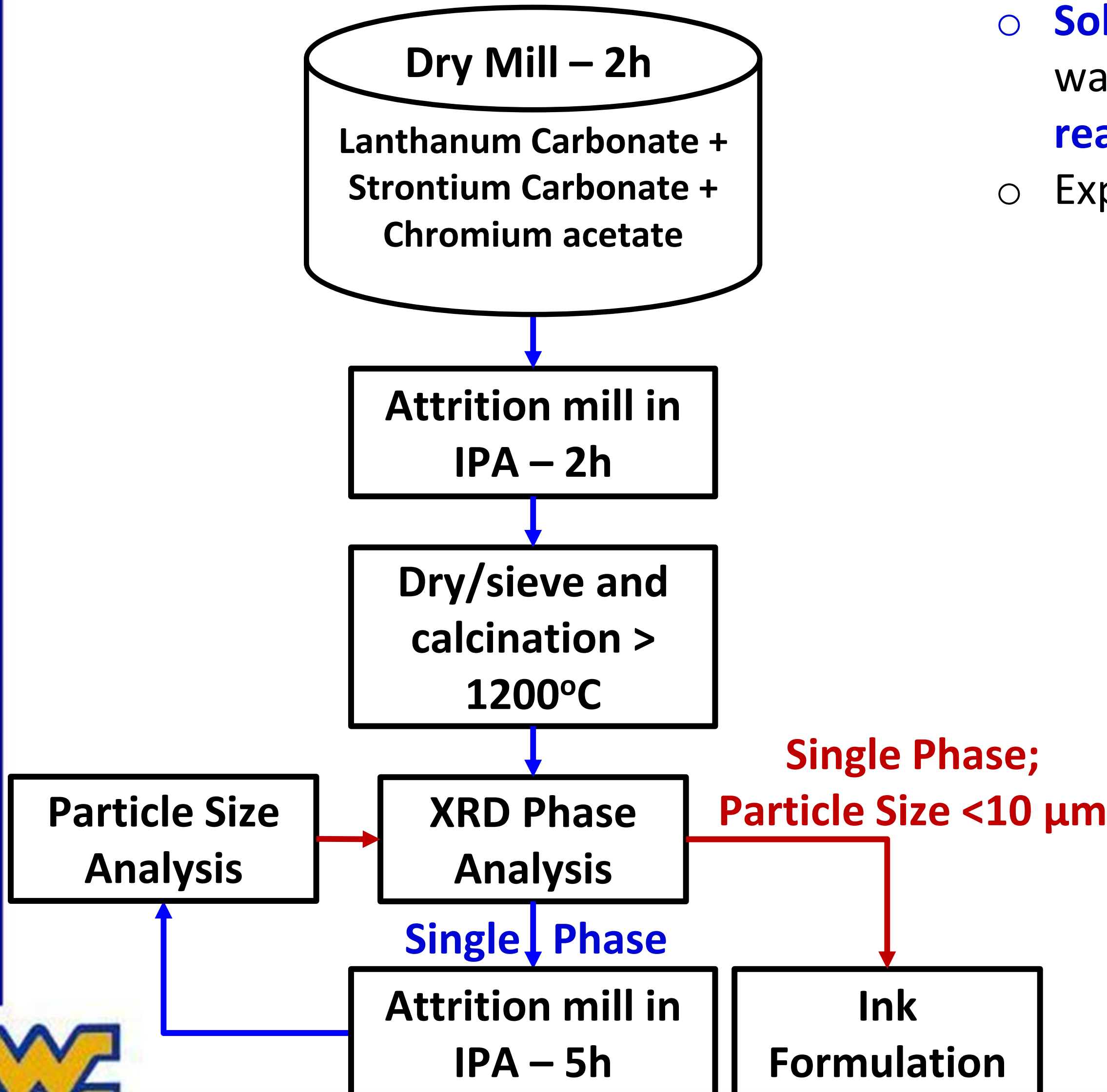
Indium Tin Oxide (ITO):

- Stable up to 1300°C.
- Electrical conductivity:>60 $S \cdot cm^{-1}$.
- Thermal stability was studied in this work.
- Good adhesion on to the substrate without the formation of secondary phase.

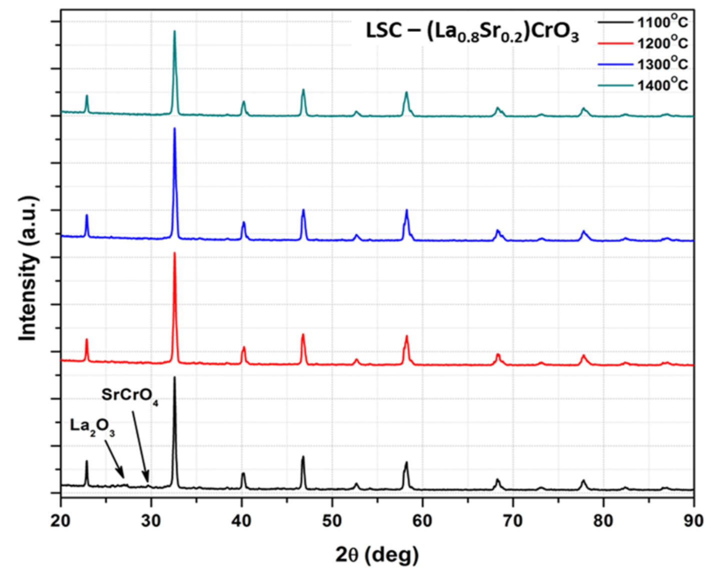
Strontium-doped Lanthanum Chromite (LSC):

- Stable up to 1800°C.
- Electrical conductivity:>3 $S \cdot cm^{-1}$.
- Thermal stability was studied in this work.
- Phase formation and compatibility with alumina substrate studied in this work.

Electroceramic Material Synthesis & Processing - LSC

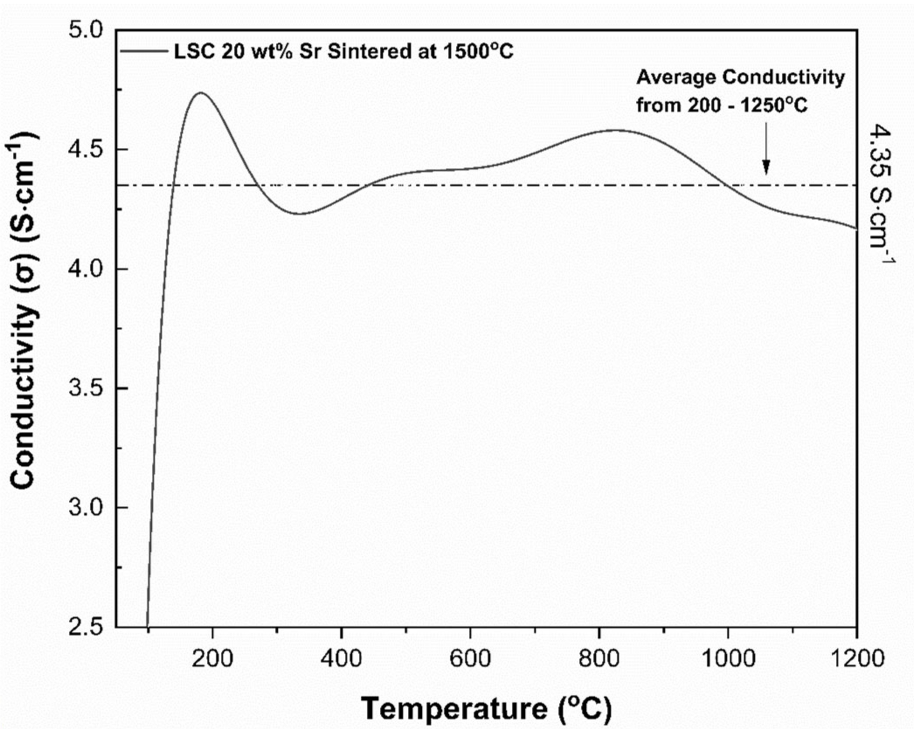


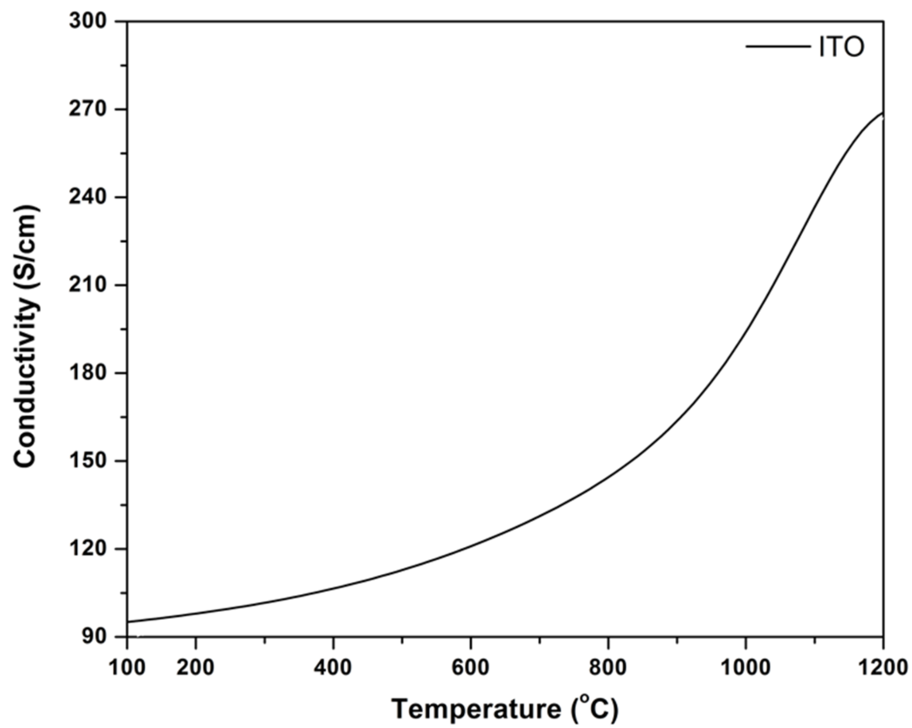
- **Solid solution** of LSC with **20% Sr** was synthesized by a **solid-state reaction** process.
- Expected phase: **$(La_{0.8}Sr_{0.2})CrO_3$**



Electrical Conductivity Analysis

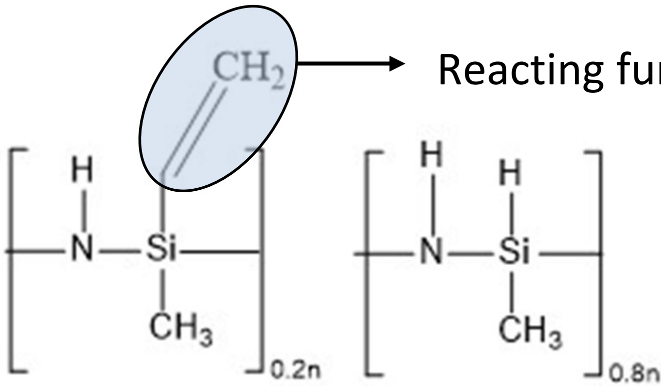
- Thick film conductivity (σ) with **thickness ~20 µm** was tested from **100 – 1250°C** for both **LSC** and **ITO**.
- $\sigma_{(LSC)}$ = ~4.34 S·cm^{-1} from 200 – 1250°C.
- $\sigma_{(ITO)}$ = ~100 - 280 S·cm^{-1} from 200 – 1250°C.





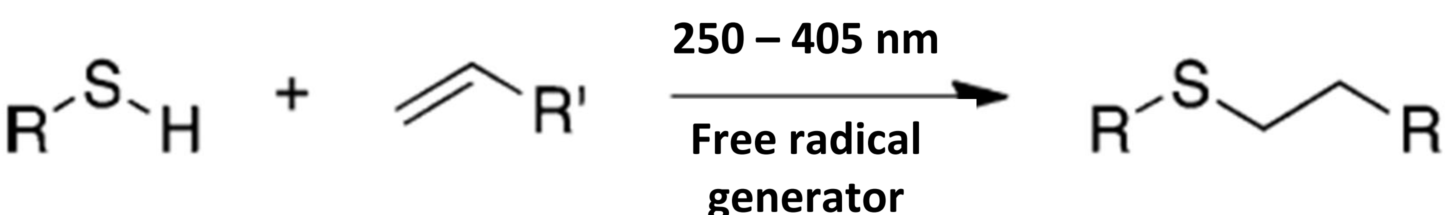
Polymer Derived Ceramics (PDCs) Processing

❖ Monomer: **Durazane – 1800 (SiCN) forming polymer.**



Reacting functionality in photopolymerization

Mechanism (Anti- Markovnikov addition polymerization):



Thiol group **Alkene Group** **Cross-Linking**

Photocurable Preceramic Polymer – Synthesis

Step 1 – Stabilization:

Mix the precursors in an **ultrasonic bath** for **2h** in a closed container.

Preceramic Polymer (Durazane – 1800) + Photoinitiator (P.I.) + Photo absorber (275 – 365 nm) + Free Radical Scavenger

Step 2 – Addition of Cross-linking Compound:

Add **dithiol** before **cross-linking** under amber light.

Stir for 1h in a closed container to homogenous the photocurable prepolymer.

Weight ratio of the Reacting Compounds:

100 (D-1800) : 20 (dithiol) : 1.5 (P.I.) : 0.05 (Photo Absorber) : 0.01 (Free Radical Scavenger)

Photocurable Preceramic Polymer – Synthesis

The photo preceramic polymer was cast into the casting mold and cross-linked at 365 nm.

$\mathbf{2 \times 1}\ \boldsymbol{mm^2}$ **casting mold**

After releasing from $\mathbf{2 \times 1}\ \boldsymbol{mm^2\ mold} - \mathbf{2.118}\ \boldsymbol{g}$

Sintering @1200°C in Ar for 2 h – 1.189 g (mass loss = 0.929)

Task 2.0 Summary:

- Synthesized and characterized composites of indium tin oxide and lanthanum strontium chromate.
- ITO and LSC showed good electrical conductivity for thermoelectric applications.
- Analyzed and determined the optimum parameters to print photocurable preceramic polymer on Al_2O_3 substrates.

Task 2.0 Near-term Future Work:

- **Synthesize:**
  - PDC systems with active and inactive (Al_2O_3, ZrO_2) fillers.
  - Characterize Durazane – 1800 (a high silicon containing polymer) by adding Mo/C to synthesize SiCN, or $MoSi_2$.
  - Explore SMP-10 (high yield SiC forming preceramic polymer).
- Effects of thermal processing.
- Study phase formation.

Task 3.0:
Direct-Writing, Patterning, and Transfer of the Sensor System.
(Sierros/Sabolsky/Varadharajan)

Task 3.0 Objectives:

- To define processes to direct-write through ink-jet and robo-casting the polymer-derived electroceramic composites onto oxide and polymer surfaces.
- To develop a method to transfer the pattern from an organic film to a ceramic surface and bond after thermal treatment.
- To develop a process based on photolithography to fabricate smaller sensor architectures to overcome the geometrical limitation of the direct-writing process.
- To develop a screen-printing process to transfer inductor and capacitor pattern to Al_2O_3 substrate.

Task 3.0 Approach:

Subtask 3.1 Direct-Writing/Screen-Printing Process Development:

- Develop and characterize inks within a permissible surface tension and viscosity level. Direct-writing with Nordson EFD Performus VI robo-printer.
- General process for droplet deposition, drying, and thermolysis will be defined.

Subtask 3.2 Micro-Casting Process Development:

- Develop a process to pattern micro sensor design directly on a ceramic and oxide substrate.
- Determine parameters for micro-casting including the viscosity, aspect ratio, particle size distribution and thermolysis.

Subtask 3.3 Baseline Sensor Testing and Design Optimization:

- Initial sensor configurations will be designed, with focus on temperature and strain measurements. The electrical performance testing will be completed at high-temperature (500-1700 ^{o}C).

Subtask 3.4 "Peel and Stick" Development:

- Investigate methods to transfer the sensor circuit/system to the active energy system component, which will be represented by alumina and zirconia substrates.

Task 3.0 Current Status:

Subtask 3.1 Direct-Writing Process Development:

- ✔ Developed inks within a permissible surface tension and viscosity level.
- ✔ Direct-writing with Nordson EFD Performus VI robo-printer and screen-printing.

Subtask 3.2 Micro-Casting Process Development:

- ✔ Developed a micro-casting process based on photolithography to pattern reduced geometry sensor structures.
- ✔ Including methods to alter the wetting and drying characteristics of the deposited composite solutions.

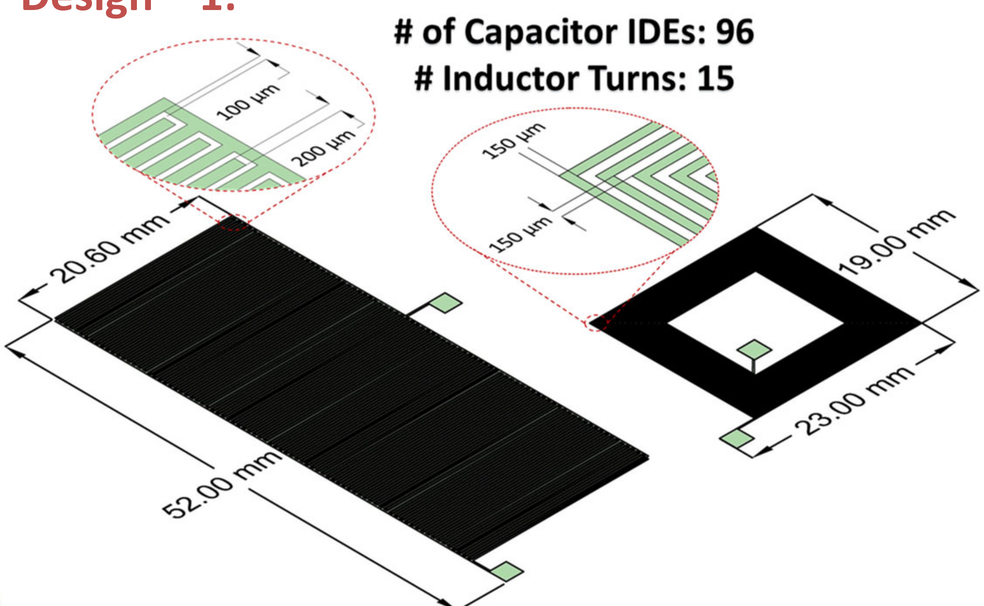
Subtask 3.3 Baseline Sensor Testing and Design Optimization:

- ✔ Ink deposited directly on oxide surfaces undergo thermal treatment defined by thermal schedules in Task 2 as a starting point.

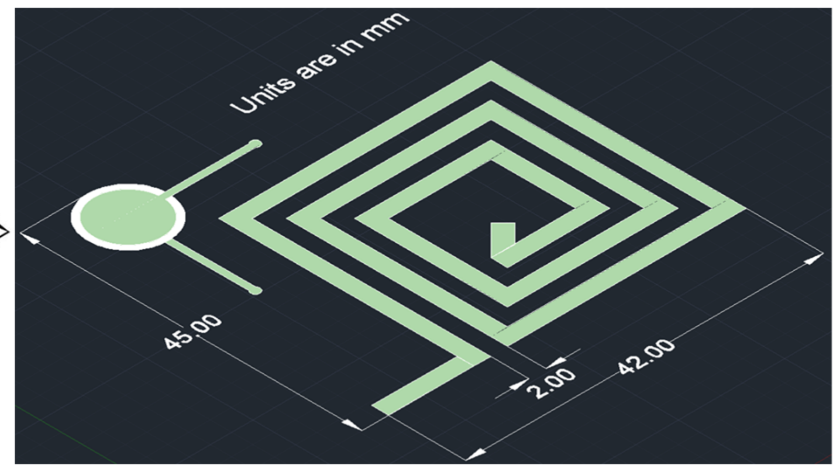
LC Resonator Design and Simulation

- Two architectures were developed using **ANSYS/CAD** drawing software based on the working frequency range simulated by **Microwace AWR**.
- **Design 1:** 2-inch inductor/**capacitor (IDC)** pattern (fabricated by **micro-casting, direct ink writing**).
- **Design 2:** 1-inch inductor/**parallel plate capacitor** (fabricated by **screen-printing**).

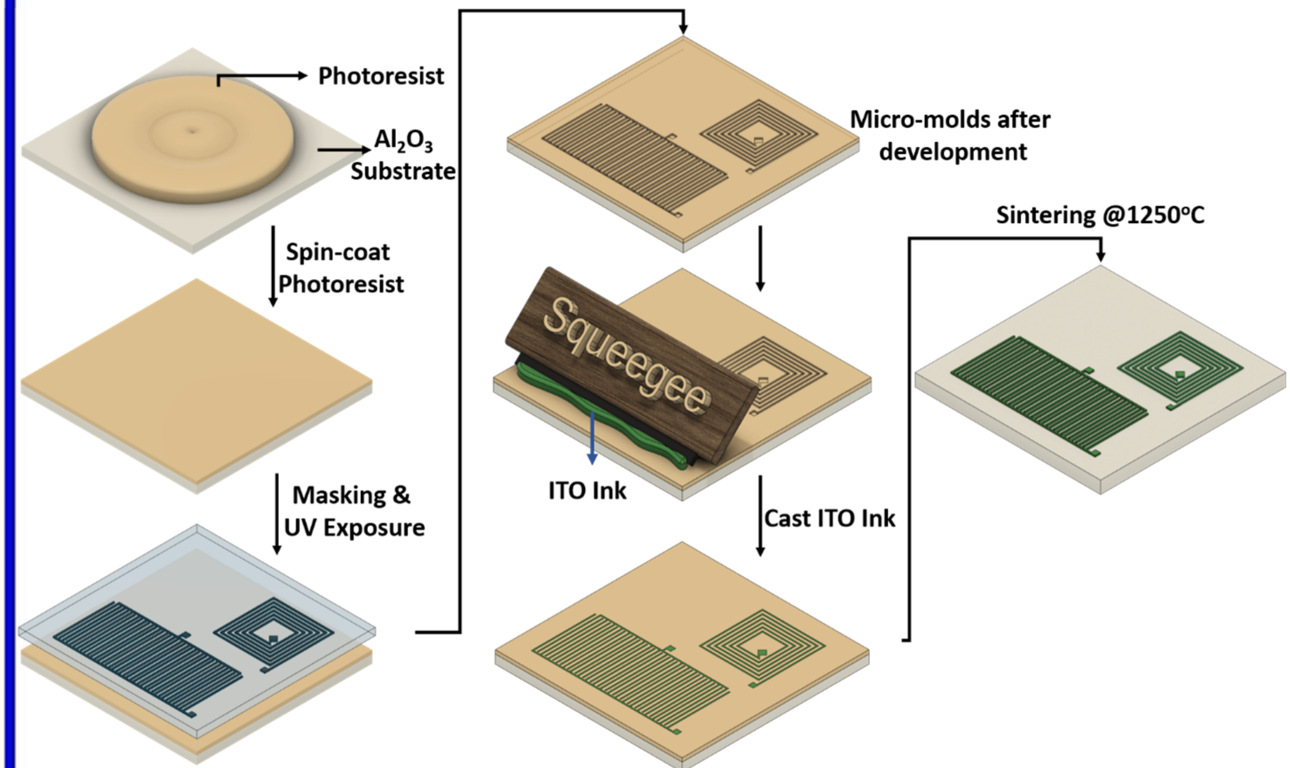
Design – 1:



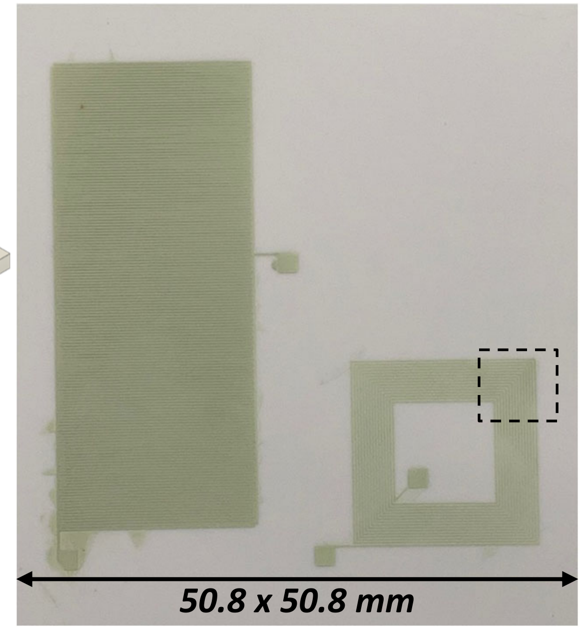
Design – 2:

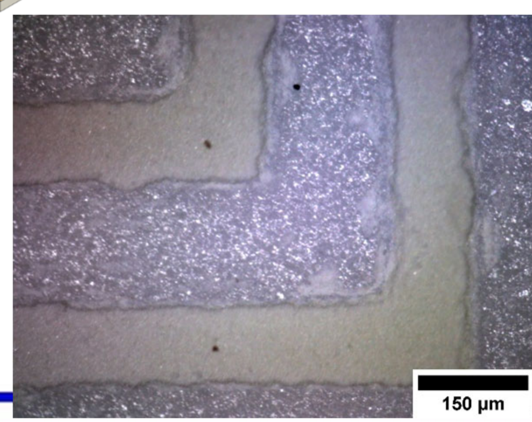


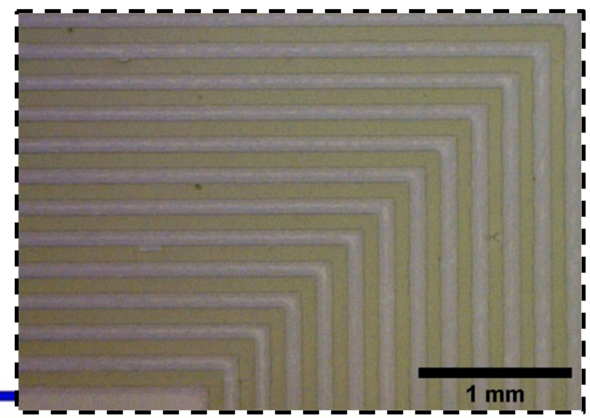
Design – 1: Photolithography & Micro-Casting Process:



50.8 x 50.8 mm sensor design on Al_2O_3 substrate.





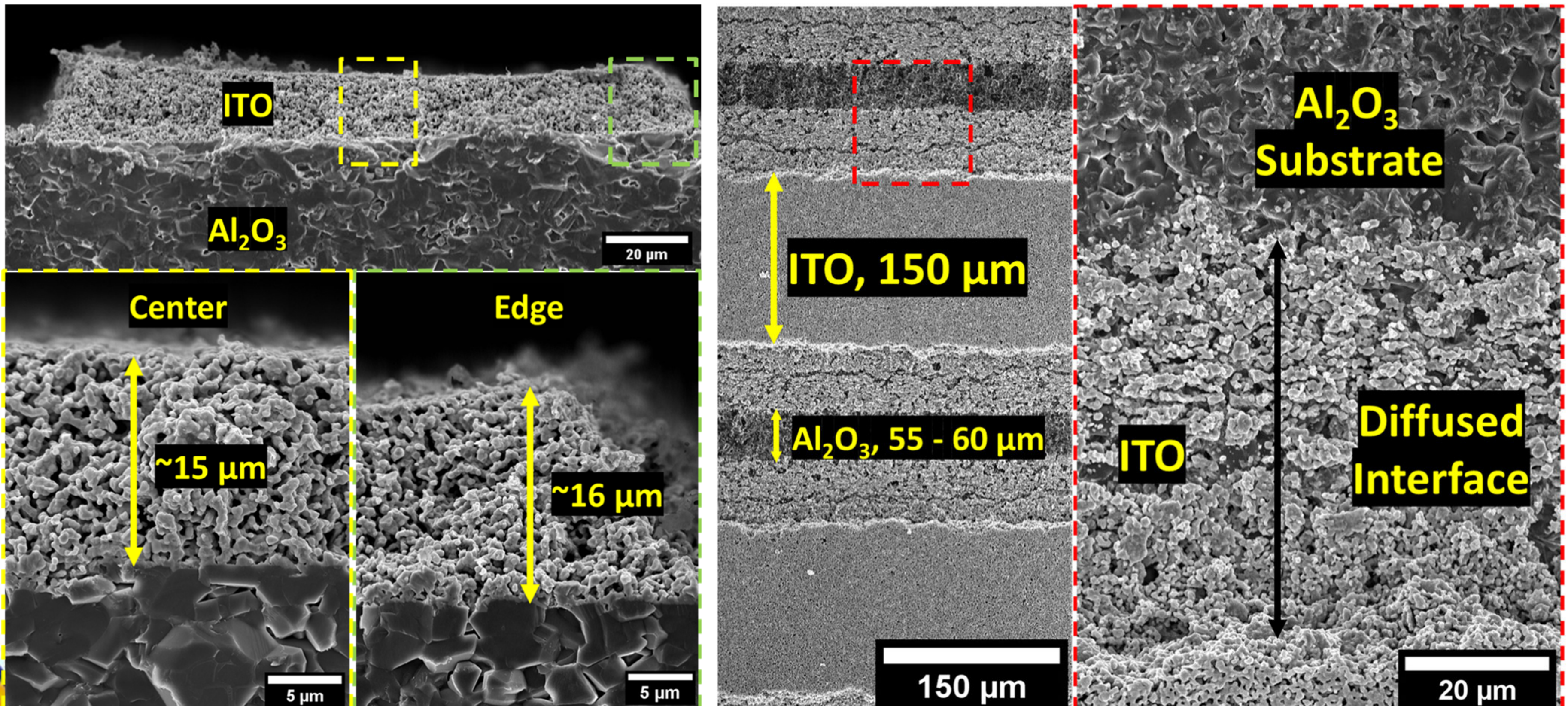


Design – 1: Microstructural Analysis

SEM micrographs of Fingers IDE Capacitor shows the effect of isotropic etch.

Isotropic Etch → Increased IDE Width → Increased Capacitance

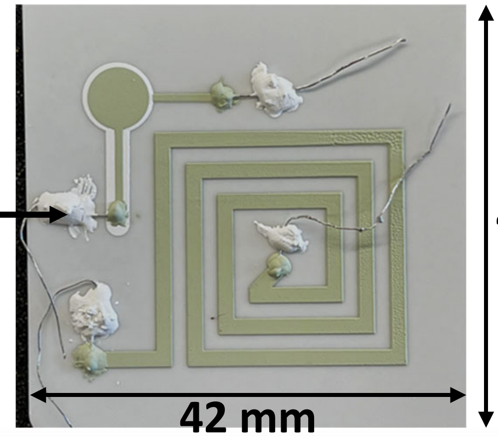
Cross-section **Topography**



Design – 2: Macro Inductor & Parallel Plate Capacitor

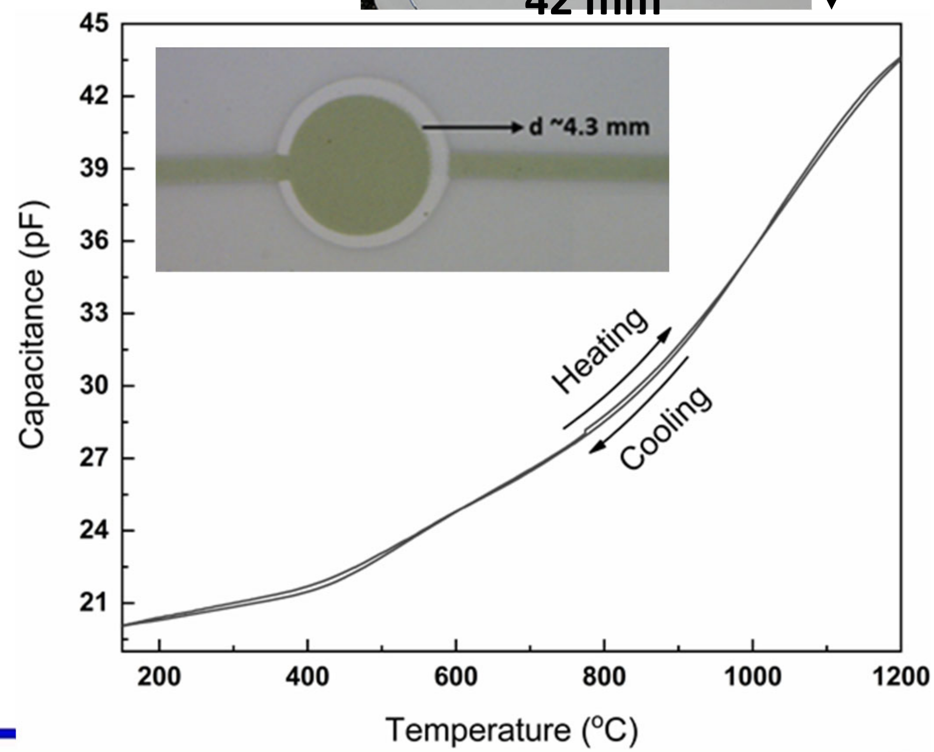
- Top & Bottom electrode: **3 layers of ITO (~60 µm)**
- Dielectric layer: **3 layers alumina (~60 µm)**

Al_2O_3 Cement Reinforcement



- The **full ceramic parallel plate capacitor** was tested to **understand** the capacitive response as a function of temperature (**C vs T**).
- **ΔC = ~2 pF/100°C.**
- The **ΔC** remains the **same** during the **heating/cooling** cycles.

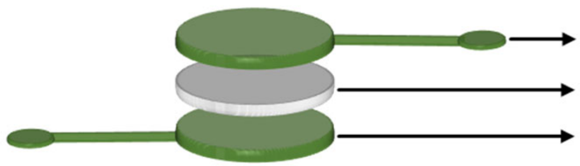
Since the C does not show hysteresis, the wireless response will not have hysteresis.

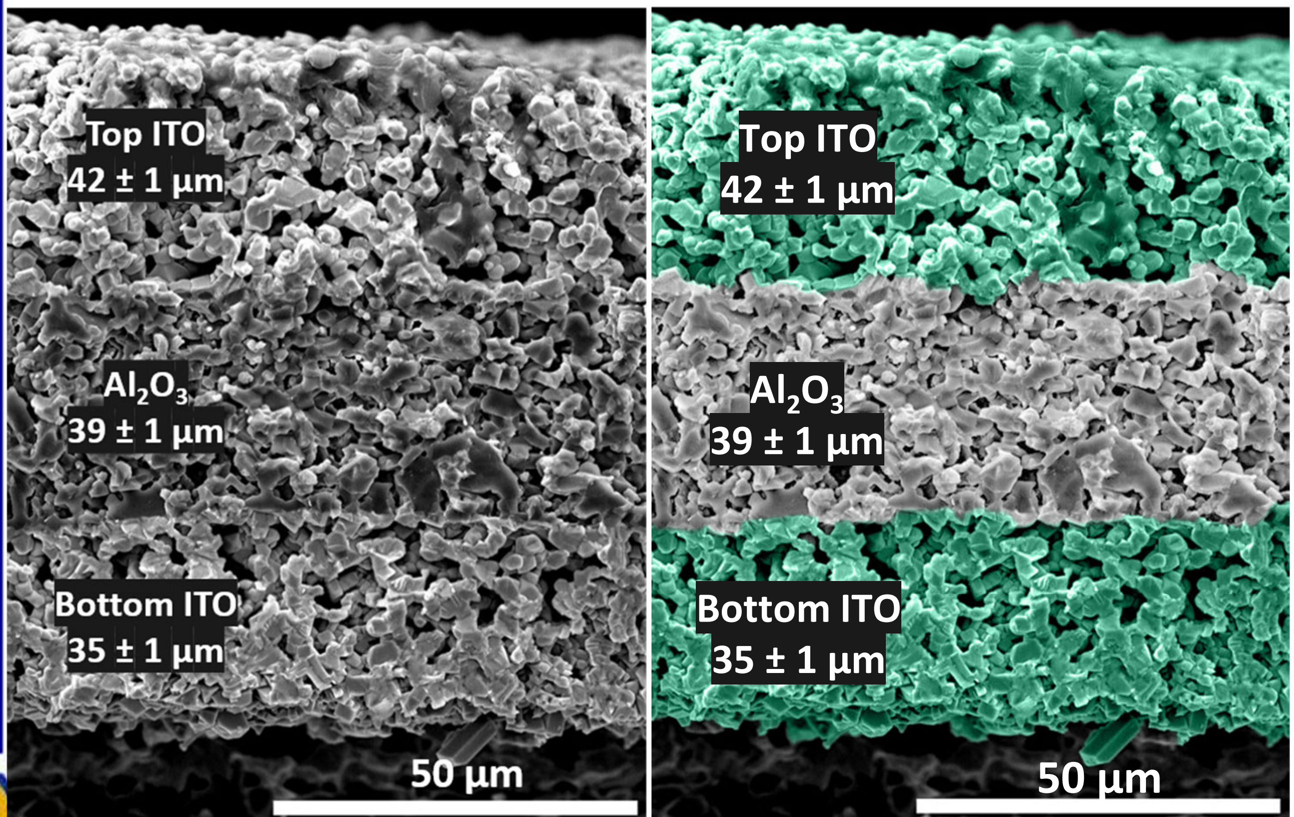


Design – 2: Microstructural Analysis – Parallel Plate Capacitor

All the layers were **screen-printed** and **co-sintered** at **1250°C** for **4 h**.

Schematic of Parallel Plate Capacitor





C @2 MHz: **~21 pF**

Dimension:
ε_r (Al_2O_3) = 9.6 (at 25°C)
A = 16 µm^2
d = 42 µm (Modelled);
39 ± 1 µm (actual)

Task 3.0 Summary:

- Micro-casting process followed to fabricate smaller form factor sensors: 2 – inch and 1 – inch designs (parallel plate) for wireless testing and evaluation.
- Microstructural stability of the ITO sensors were evaluated for isothermal hold up to 96 h at 1050^{o}C showing no change in the grain size.
- Screen-printing was used to fabricate the parallel plate capacitor and the sensor design-2.
- Increased the width of the inductor electrodes while keeping the overall geometry within 2 inches.

Task 3.0 Near-term Future Work:

- Screen-print functional ceramic ink (ITO, LSC) onto oxide substrates and optimize firing temperature for optimum density.
- Print and analyze “fully passivated” planar inductor and parallel plate capacitor design.
- Translate the technology and print LSC on alumina substrate to carry out wireless characterization up to 1500^{o}C.
- Develop stereolithography technique to 3D print PDCs on various substrates.

Task 4.0: Passive Wireless Communication Circuit Design and Testing. (Reynolds/Pozo)

Task 4.0 Objectives:

- Design and model passive wireless sensors and interrogator antennas as RLC circuits
- Fabricate and test the sensor up to 1200^{o}C
- To extend sensor performance up to > 1500^{o}C
- Advanced materials/writing processes => existing sensing strategies does not work!
- ***Innovation:*** *advanced materials, advanced processes,* ***robust and adaptive signal processing***

Task 4.0 Approach:

- Design RLC circuits via simulation for various material properties
- Develop a robust and adaptive signal processing approach to measure temperature wirelessly. *Ink/substrate material characteristics may not be known precisely or may change with heating/cooling cycles* ***=> tracking the resonant frequency does not work!***
- **Modify circuits and signal processing as needed for advanced materials and high temperatures.**

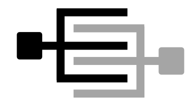
Task 4.0 Current Status:

- ✔ Completed wireless characterization of ITO (design-1) sensors from 500 – 1200^{o}C.
- ✔ New algorithms were developed to replace conventional signal processing techniques for improves data analysis.
- ✔ Replaced the conventional signal generator/network analyzer with a USRP to simultaneously transmit and receive wireless signals.
- ✔ Completed initial analysis of fully passivated LC resonators and determined the maximum working distance of electroceramic antennae.

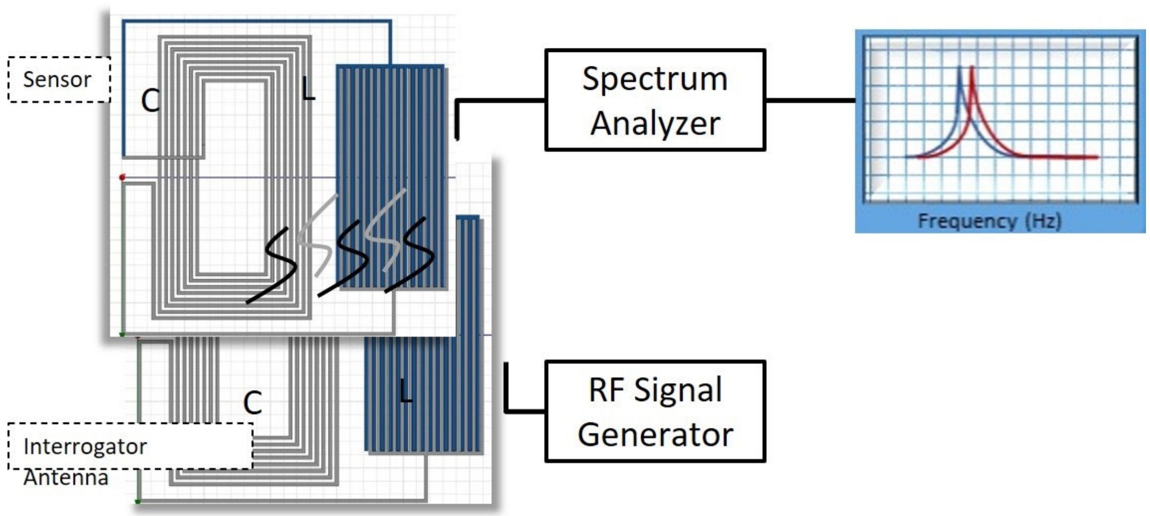
Idealized LC Passive Sensing (state-of-the-art)

- A planar inductance coil (L) and a capacitor (C) form the LC circuit and are placed on the surface of the sensor/interrogator.

Capacitor using microstrip lines (IDC)
And
Parallel Plate Capacitor →

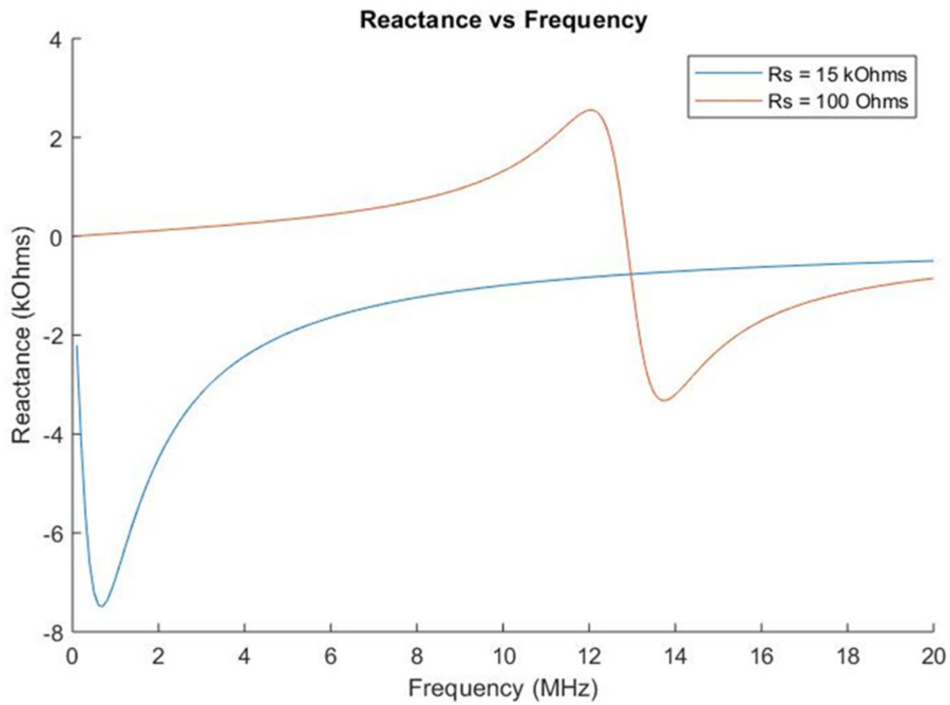


- Main idea: the capacitor is temperature dependent which causes shifts in the sensor's resonant frequency $\left(f \propto \frac{1}{\sqrt{LC}}\right)$



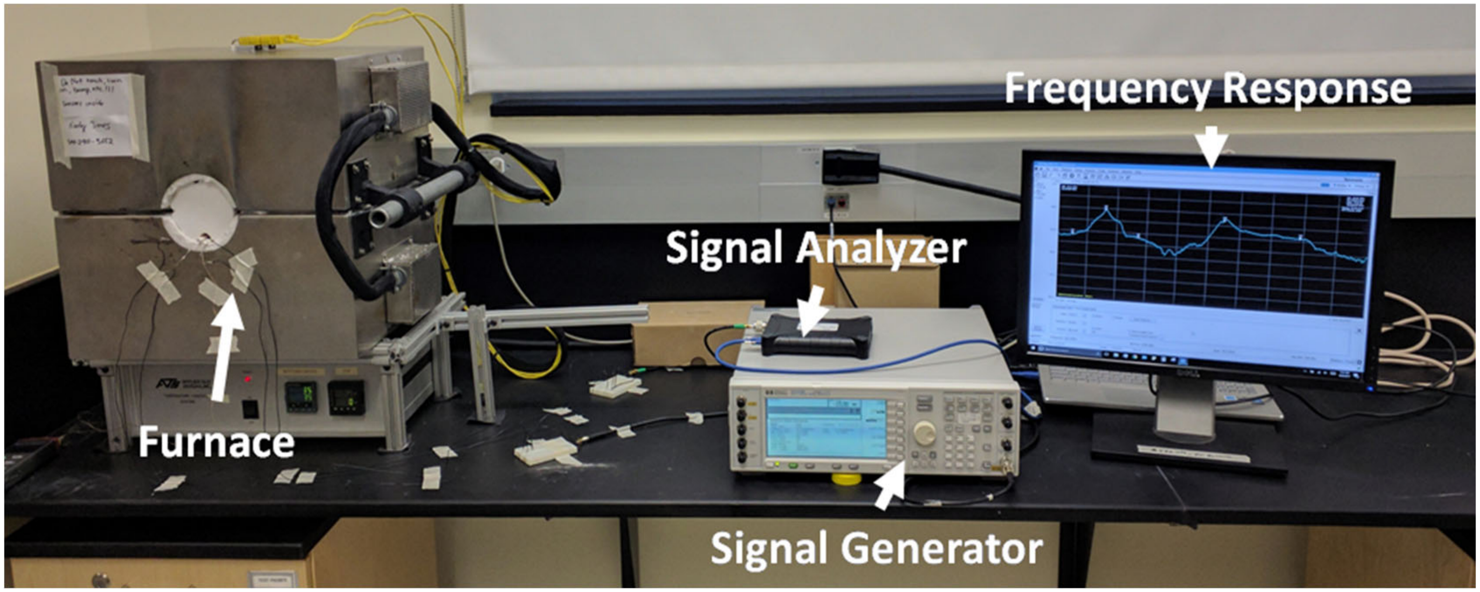
Simulation of Resistivity on Reactance vs Frequency of the Inductor

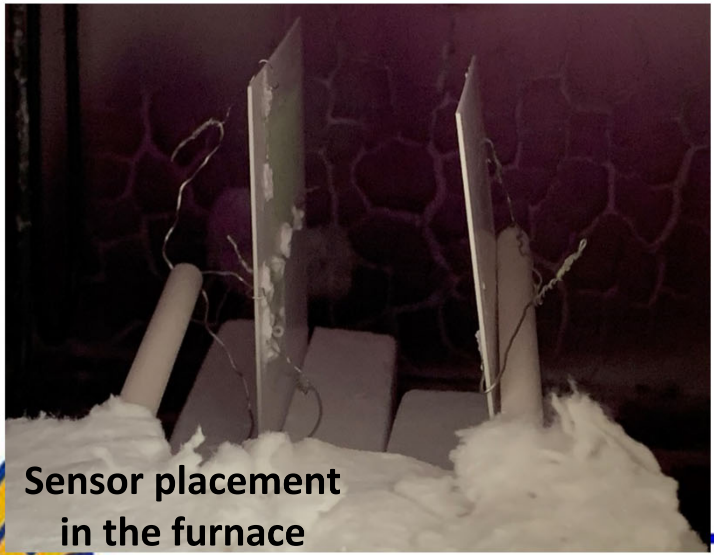
- **Reactance determines whether the component is capacitive or inductive. Inductor should have +ve reactance whereas the capacitor will show –ve reactance.**
- **High resistance of the planar inductor electrodes (15 kΩ) causes -ve reactance. (-ve reactance decreases the inductive coupling of the inductor)**
- **Lower resistance (100 Ω) shows +ve reactance at the operating frequency region.**



*D B Meadowcroft 1969, J. Phys. D: Appl. Phys. **2 1225**

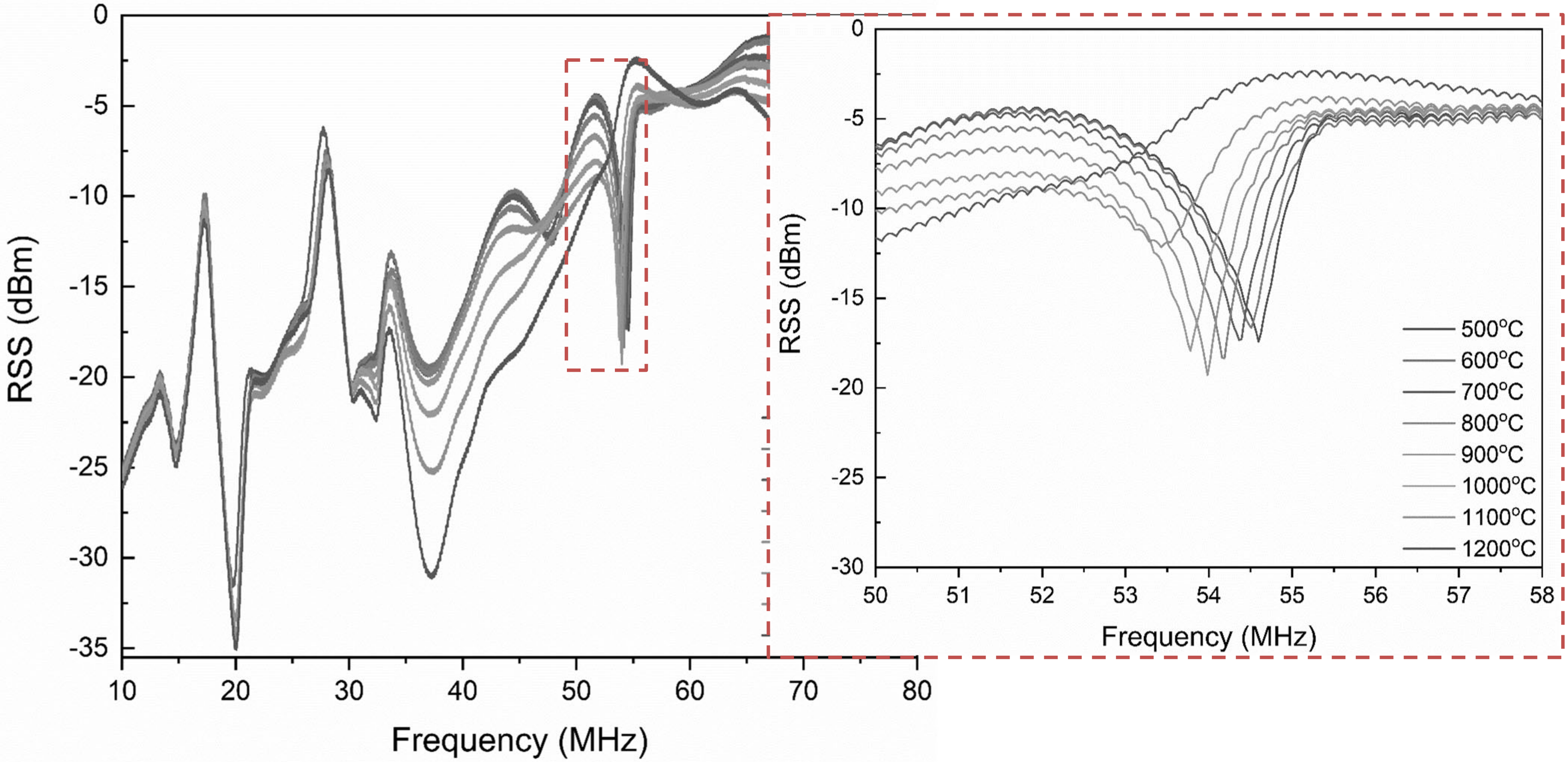
Sensor Characterization Setup





- **Two LC resonators were used to characterize the wireless signal response.**
- **One is connected to signal generator and the other to signal analyzer.**

Wireless Characterization of the Sensor Design-1: ITO Electrodes



- ❖ **Temperature signatures of the sensor from 500 – 1200°C.**
- ❖ **The peak shift can be distinguishably observed for each temperature.**
- ❖ **Temperature signatures are unique and distinguishable with a sensitivity of 3 kHz/°C.**

Frequency Analysis & Signal Processing Mechanism

❖ Data was analyzed by performing modular and piecewise comparison of frequency instead of entire spectrum.

2 algorithms were used to analyze the data:

❖ Minimum absolute Error finds the minimum difference absolute error between unknown reading data and the signatures.

❖ Cross-Correlation finds the degree of similarity between unknown reading and signatures.

Sliding Window (12 matching):

❖ Sliding-window of frequency size: 5, 10, 50, 100, 500, 1000, 2000, 2400.

❖ Example of a sliding window with a window size of 5.

✔ - Match

✗ - Not a match

Signature	10.0000	10.0292	10.0583	10.0875	10.1167	10.1458	10.1750	10.2042	10.2333	10.2625
Unknown	10.0000	10.0292	10.0583	10.0875	10.1167	10.1458	10.1750	10.2042	10.2333	10.2625

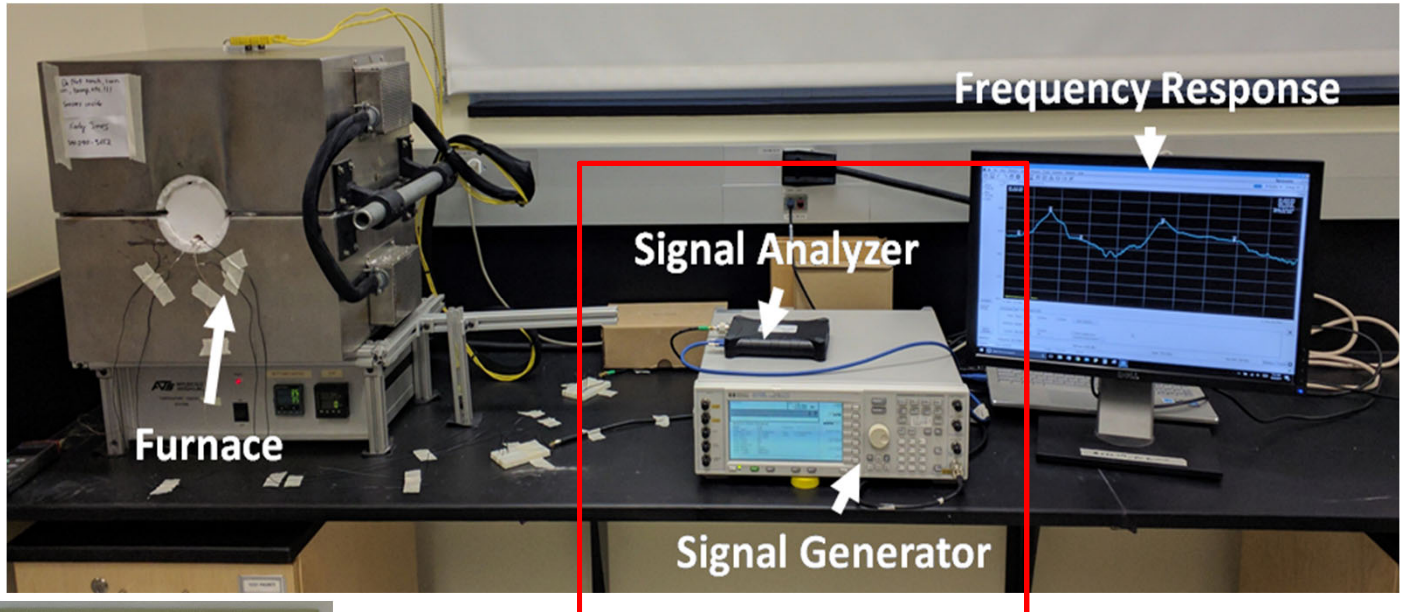
❖ Optimal window size – 500 : 50 to 58 MHz

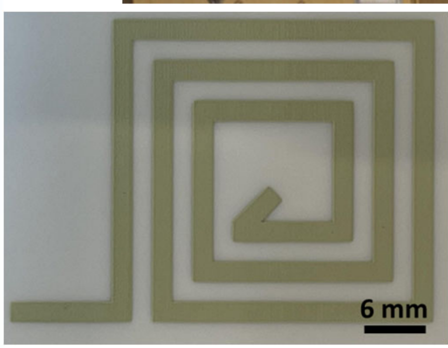
500°C- ✔ ✔; 600°C - ✔ ✔; 700°C- ✔ ✔; 800°C- ✔ ✔; 900°C- ✔ ✔; 1000°C- ✔ ✔

Software Defined Radio

- Universal Software Radio Peripheral (USRP) is a software defined radio designed by Ettus research
  - Small, cheap alternative to spectrum analyzers and signal generators
  - Open-source software, existing libraries in open source (GNU Radio), Simulink (Matlab).

USRP Usage in Current Setup



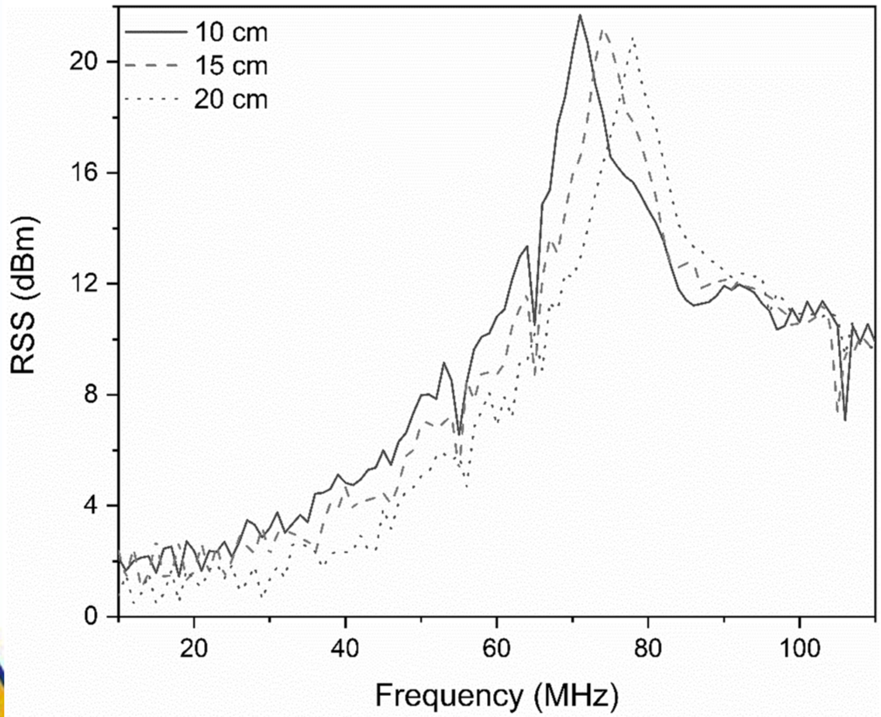


Conventional PCB Antennae was **replaced** with **ceramic antennae.**

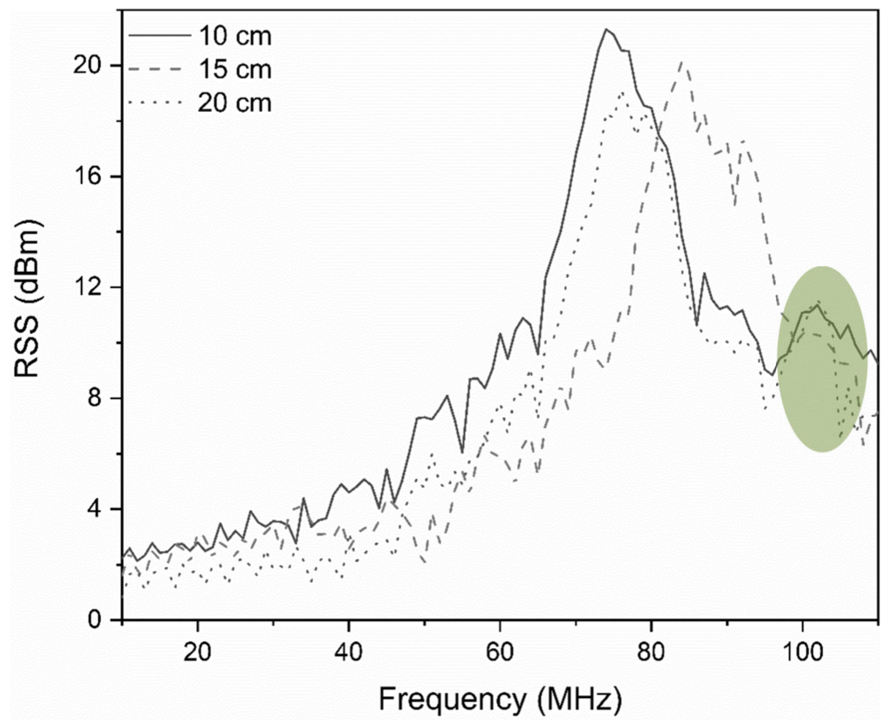
Wireless Characterization of the Sensor Design-2: USRP

- **USRP used to receive and transmit wireless signal using electroceramic antenna.**
- **Placing the LC resonator b/w the antenna showed a peak at ~102 MHz, which is the corresponding self-resonance.**

ITO Antennae Only



With LC Resonator b/w Antennae

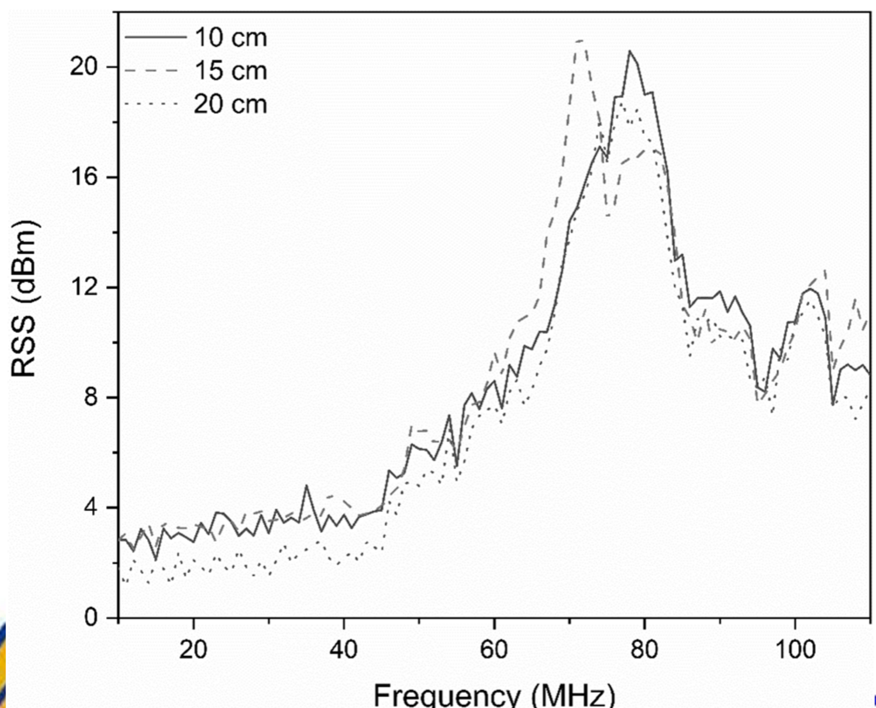


Thru' Ceramic Wireless Characterization

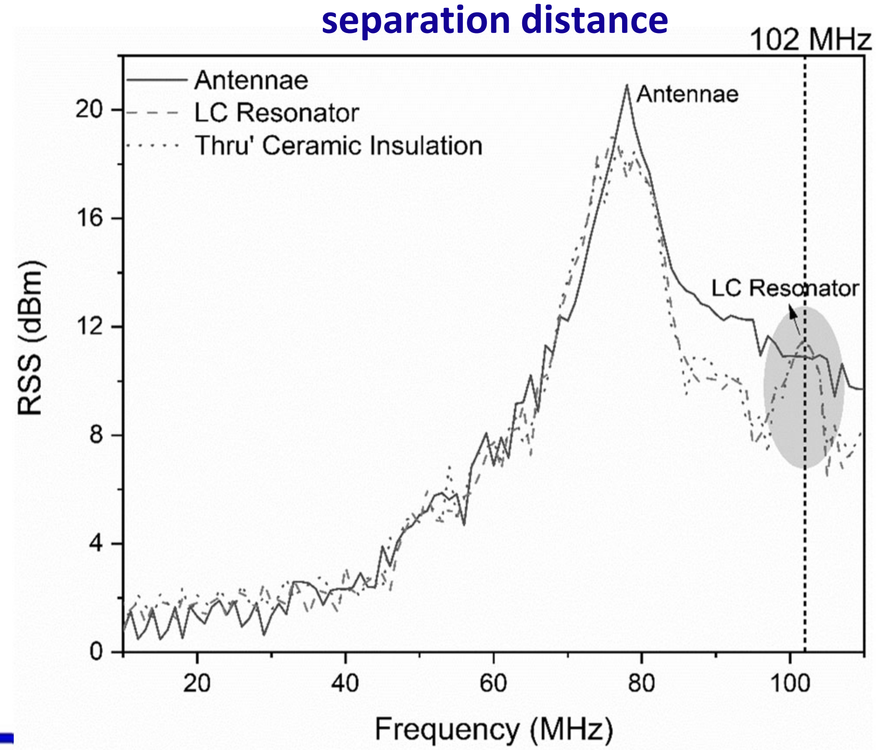
- **A ceramic (Al_2O_3) brick of ~3-inches thickness placed b/w the LC resonator and the antennae to test the signal attenuation.**

- **The Al_2O_3 ceramic does not show any significant attenuation in the wireless signal.**

Wireless Signal Thru' Ceramics at Distance 10 – 20 cm



Comparison of Wireless Signal at 20 cm separation distance



Task 4.0 Summary:

- Completed high temperature passive wireless temperature sensing
  - Created a high temperature sensing testbed.
  - Successfully demonstrated that the sensor can serve as a high temperature passive wireless sensor.
  - Developed an alternative to conventional wireless testing setup for fast and efficient real-time signal processing using USRP.

Task 4.0 Near-term Future Work:

- The sensor will be made truly passive by placing them within the range of the transmitter and the receiver.
- High temperature characterization using USRP powered wireless sensing platform.

Acknowledgments:

- We would like to thank **U.S. Department of Energy (DOE)** for sanctioning this project **DE-FE0026171**.
- **Jessica Mullen**, project manager at U. S. Department of Energy, is greatly appreciated for her insight and valuable guidance.
- We also would like to acknowledge Mr. Harley Hart, Dr. Qiang Wang, and Dr. Marcela Redigolo for their cooperation and valuable assistance in the WVU Shared Facilities.
- Kindly acknowledge faculty and staff of West Virginia University for their support.

Thank you!!!

Questions

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