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

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Background- Harsh Environment Sensing Needs

- Online monitoring of energy systems in extreme conditions is required for mining/drilling, transportation, aviation, energy, chemical synthesis, and manufacturing applications.
- Harsh-environments include:
  - High temperature (1000^{o}C-2000^{o}C).
  - High pressure (up to 1000 psi).
  - Various pO_2 levels.
  - Corrosive conditions (molten inorganics or reactive gasses) .
- Ability to monitor:
  - Temperature
  - Stress/strain within energy or reactor components
  - Failure events
  - Overall health

Processing Vision – Peel and Stick Sensor

Item A represents the organic carrier film.

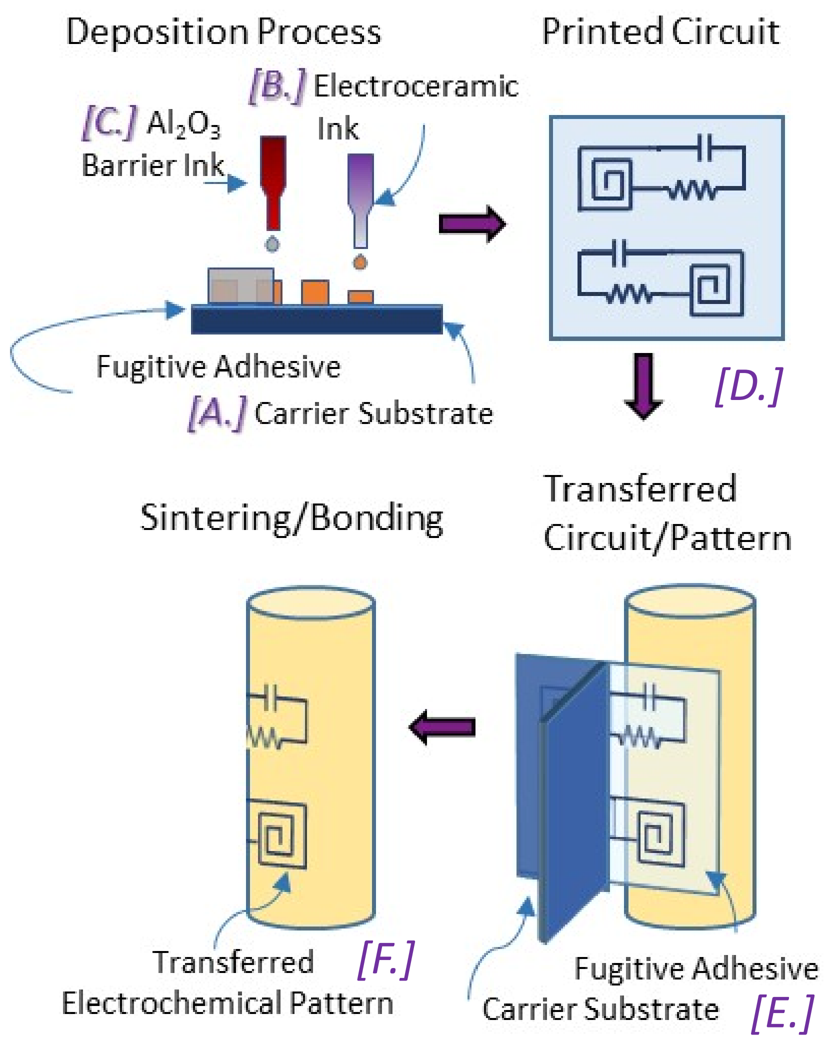
Item B represents the polymer-precursor ink (converts to an electroceramic after heat treatment).

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 electroceramic 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.

R&D Team (Co-PIs)

Dr. Edward M. Sabolsky (WVU Mechanical and Aerospace Engineering) will act as PI of the program (both technical and administrative), and will be responsible for ceramics processing and sensor testing.

Dr. Kostas Sierros (WVU Mechanical and Aerospace Engineering) will lead development of micro-patterning and robo-casting of ceramic materials, and will be the co-developer of the printing inks and direct-writing tasks.

Dr. Daryl Reynolds (WVU Computer Engineering) will lead the electronics design, interfacing and circuitry, in addition to the development of the passive wireless communication and testing.

Dr. Matthew Seabaugh and Mr. Gene Arkenburg (Nexceris LLC) are Director of Product Development and SOFC Group Leader, respectively. Assist in testing technology on SOFC platform.

Dr. Kristen Brosnan (GE Global Research) is the Manager of Ceramic Structures and Processing Laboratory. Consult and mentor team for turbine blade application and demonstration.

Major Milestones:

Materials/Sensor Fabrication

M1- (Task 2)⇒ Down-select composite composition for Task 3. (June 2016→June 2018).
M2- (Task 3)⇒ Define basic ink/paste formulation for printing. (June 2016→Aug. 2018).
M3- (Task 3)⇒ Completed baseline sensor printing on oxide substrates. (August 2017).
M4- (Task 3)⇒ First demo of pattern transfer. (Oct. 2018→Oct. 2019).
M5- (Task 3)⇒ First demo of circuit pattern transfer. (March 2017 →Nov. 2019).

Passive Wireless Circuit Modeling and Testing

M6- (Task 4)⇒ Completed design and testing of sensor circuit. (Sept. 2016).
M7- (Task 4)⇒ Completed wireless coupling modeling for applications. (March 2017).
M8- (Task 4) ⇒ Establish high-temp testing setup. (Nov. 2017).

- Michael Comparetto (M.S. student)- sensor testing (**Graduated** Dec. 2018)
- Kavin Sivaneri Varadharajan Idhaiam (Ph.D. student)- materials/sensor research (started Jan. 2018).
- Harish Palakurthi (Ph.D. student)- electronics/wireless research (started Jan. 2018).

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:** La_2NiO_4 composites
- **System 2a:** Indium tin oxide (ITO)
- **System 2b:** Lanthanum strontium chromate (LSC)

Task 2.0 Approach:

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

Electrically conducting/semi-conducting oxides namely, lanthanum nickelate (La_2NiO_4), 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^{o}C.

- **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:

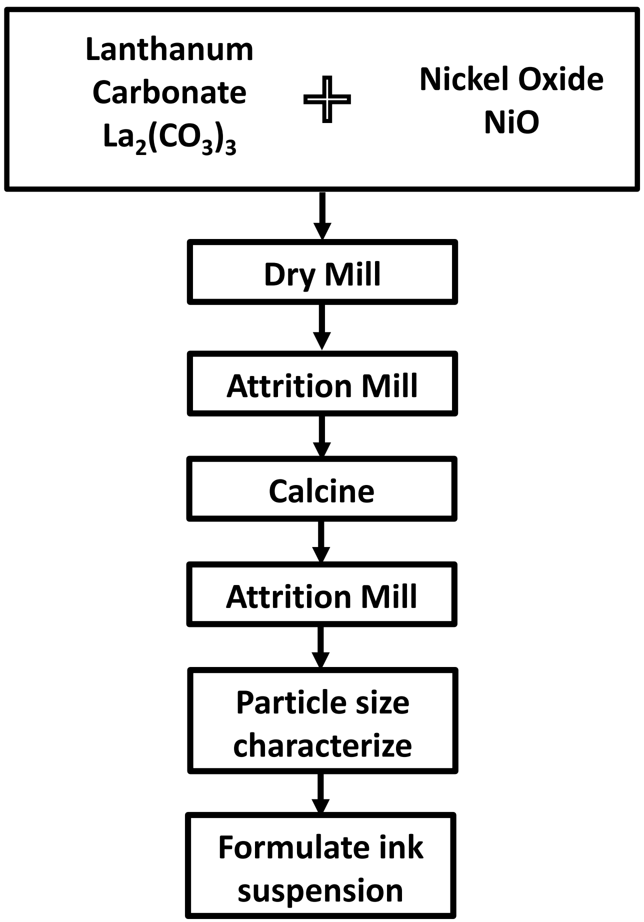
- **Subtask 2.1 Synthesis of Multifunctional Electroceramic Composites:**
  - ✓ ***System 1:*** La_2NiO_4-Al_2O_3 composites.
  - ✓ ***System 2a:*** Indium tin oxide (ITO).
  - ✓ ***System 2b:*** Lanthanum strontium chromate (LSC).

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

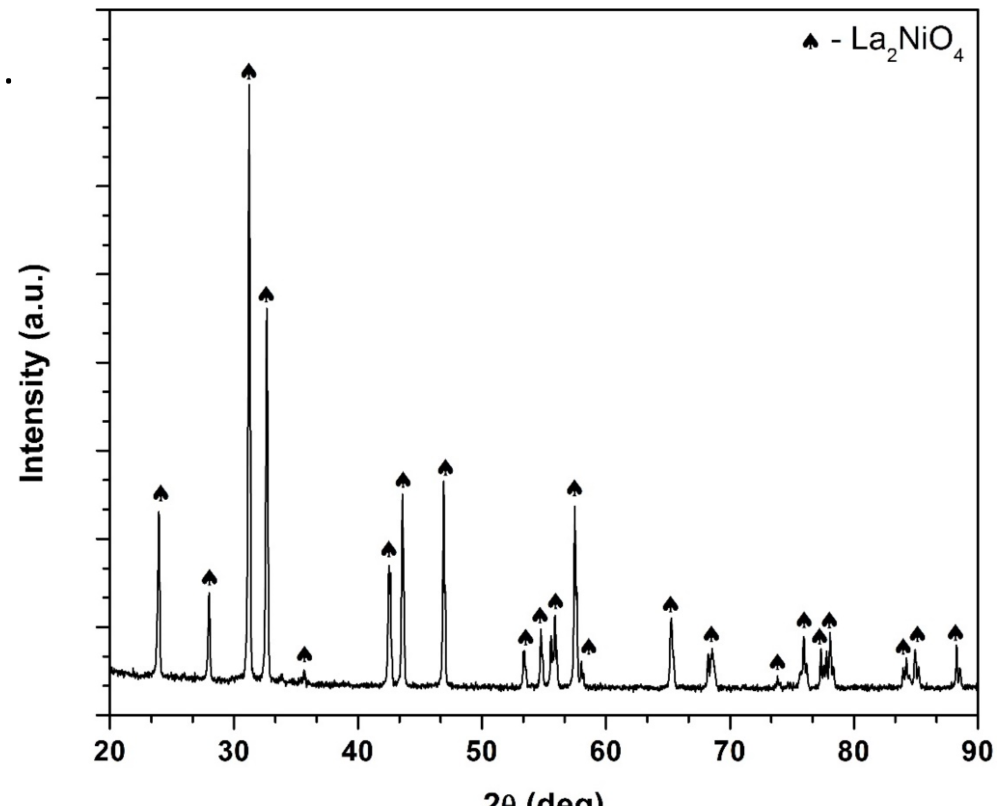
- **Subtask 2.3 Composite Material Testing and Characterization:**
  - ✓ Phase/chemistry characterization is completed on the oxide electroceramic composites.

System 1: Conductive oxide- Lanthanum Nickelate

- P-type semiconductor (some mixed-conduction).
- CTE~ 13×10^{-6} /K (with K_2NiF_4 structure).
- σ = 60-100 $S\cdot cm^{-1}$ (200-1400^oC)*.



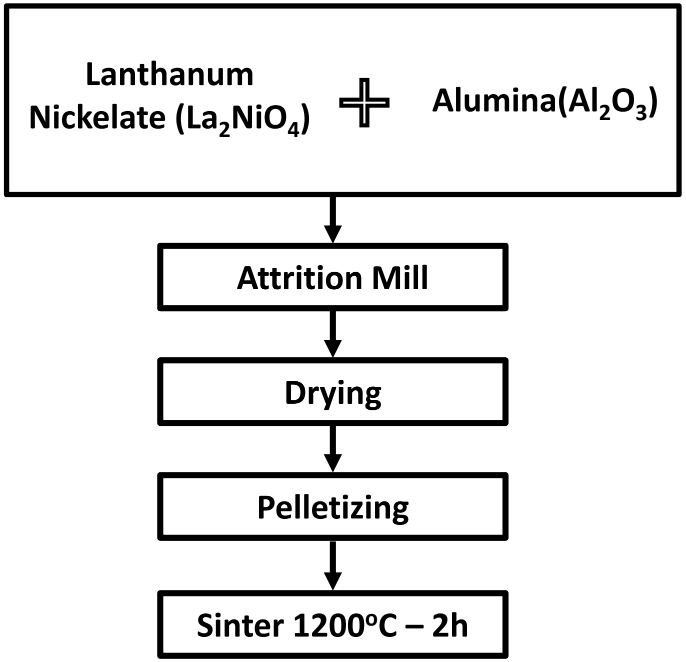
X-Ray Diffraction of Lanthanum Nickelate



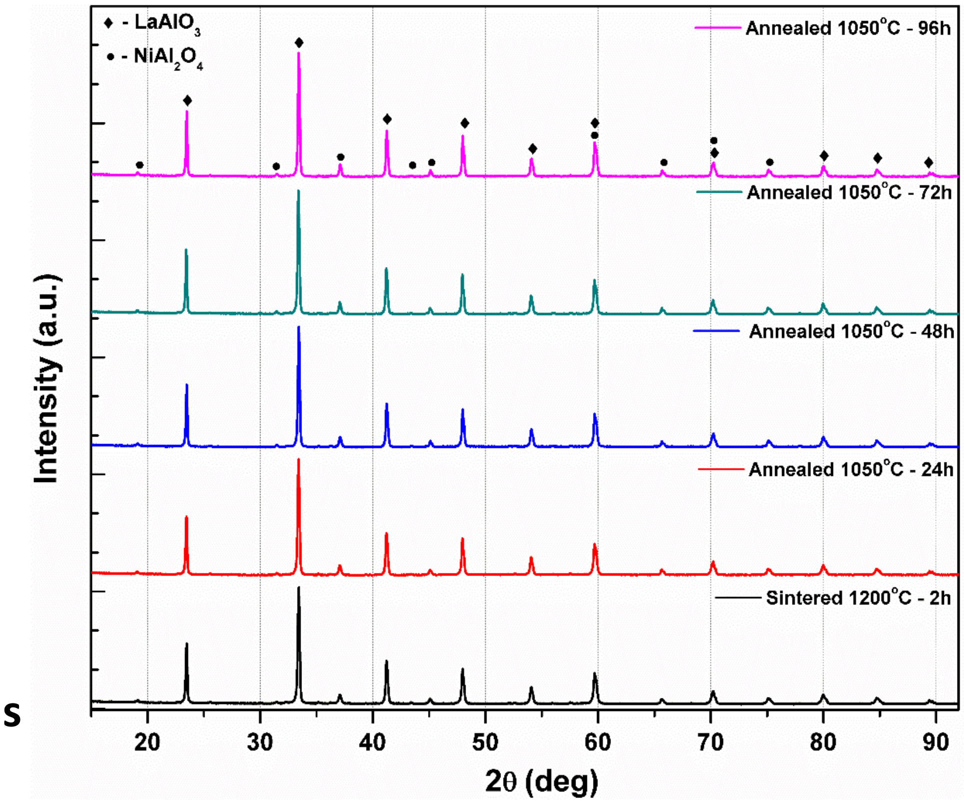
❖ **Due to the delamination of lanthanum nickelate on alumina (Al_2O_3) substrate, composite of La_2NiO_4-AL_2O_3 was synthesized and characterized.**

*Ceramics International 34 (2008) 651–655

Fabrication and Characterization of 50-50 vol.% La_2NiO_4 – Al_2O_3 Composite



X-Ray Diffraction 50-50 La_2NiO_4 – Al_2O_3



- **50-50 La_2NiO_4 – Al_2O_3 composite forms $LaAlO_3$ and $NiAl_2O_4$ phases at 1200ºC.**
- **Rietveld refinement shows the phases are stable after initial phase transformation.**
- **After annealing, the density of the composite was increased by ~3%.**

Phase	1200ºC – 2h (Wt.%)	1050ºC – 96h (Wt.%)
$LaAlO_3$	51.64	49.90
$NiAl_2O_4$	48.36	50.10

Fabrication and Characterization of 60-40 vol.% La_2NiO_4 – Al_2O_3 Composite

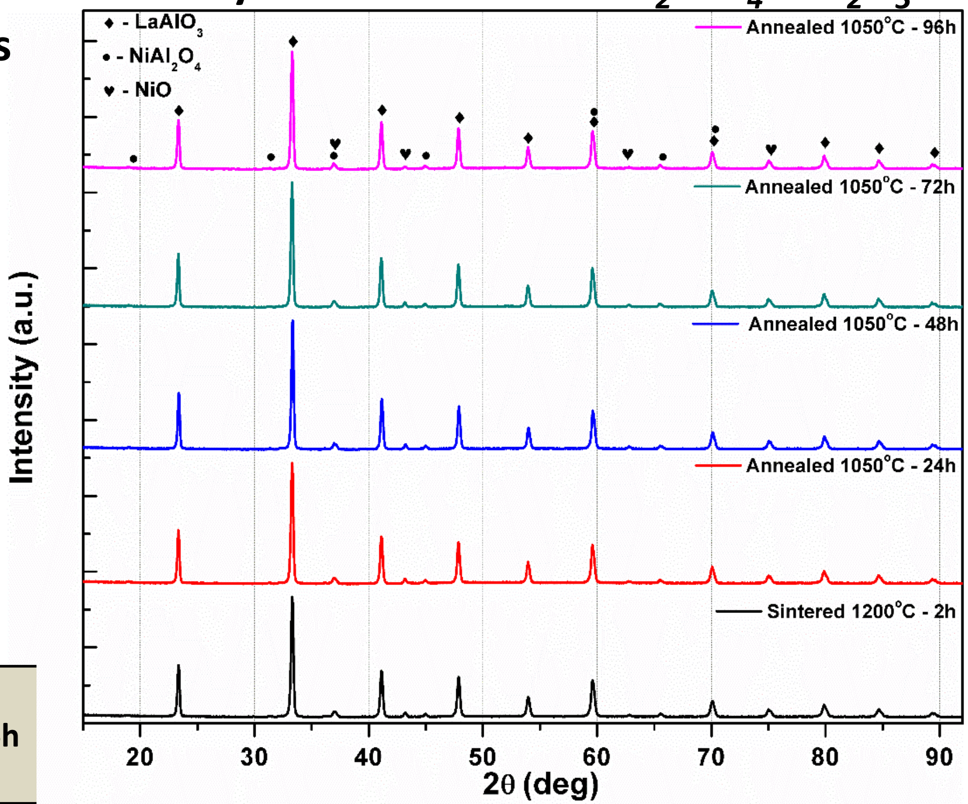
- **60-40 La_2NiO_4 – Al_2O_3 composite forms $LaAlO_3$, $NiAl_2O_4$, and NiO phases at 1200°C.**

- **The $LaAlO_3$, $NiAl_2O_4$, and NiO phases remain stable over the annealing temperature.**

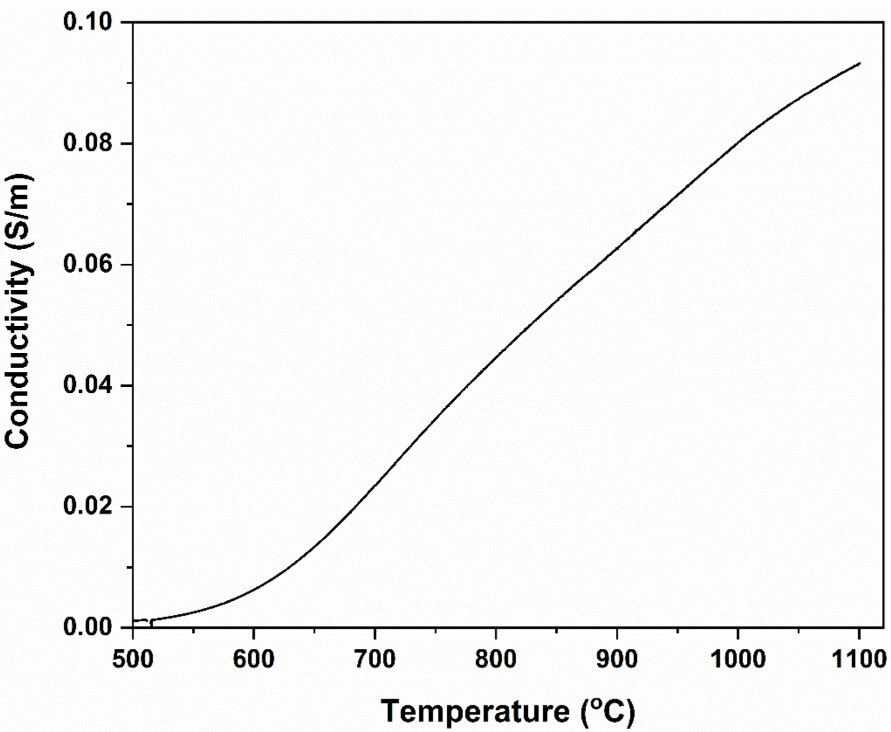
- **During annealing, the density of the composite was increased by ~4.9%.**

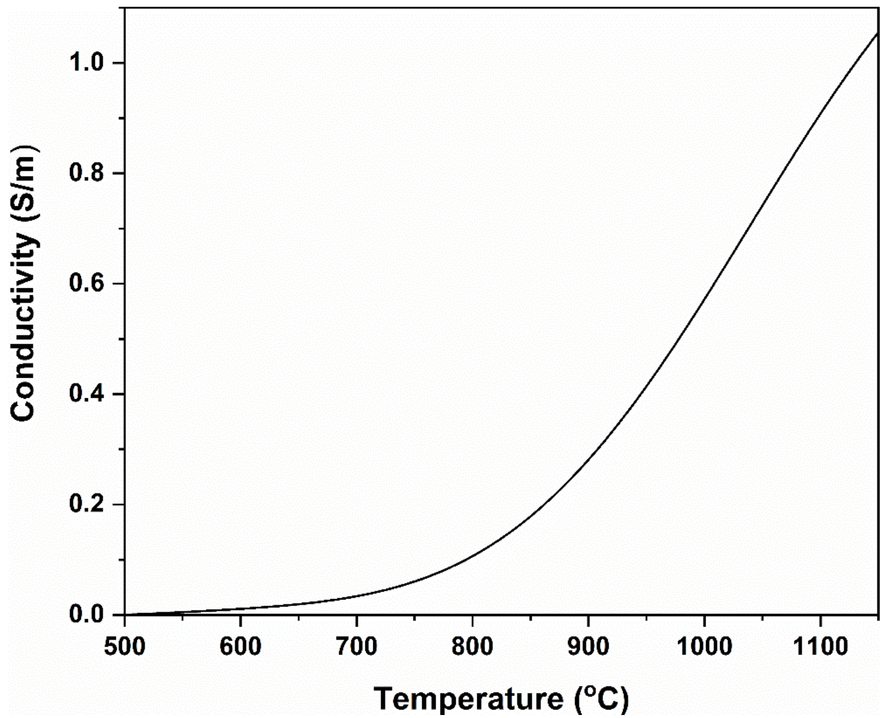
Phase	1200°C – 2h (Wt.%)	1050°C – 96h (Wt.%)
$LaAlO_3$	36.30	38.71
$NiAl_2O_4$	30.84	31.27
NiO	32.86	30.02

X-Ray Diffraction 60-40 *La_2NiO_4 – Al_2O_3*



Electrical Conductivity of the Composites

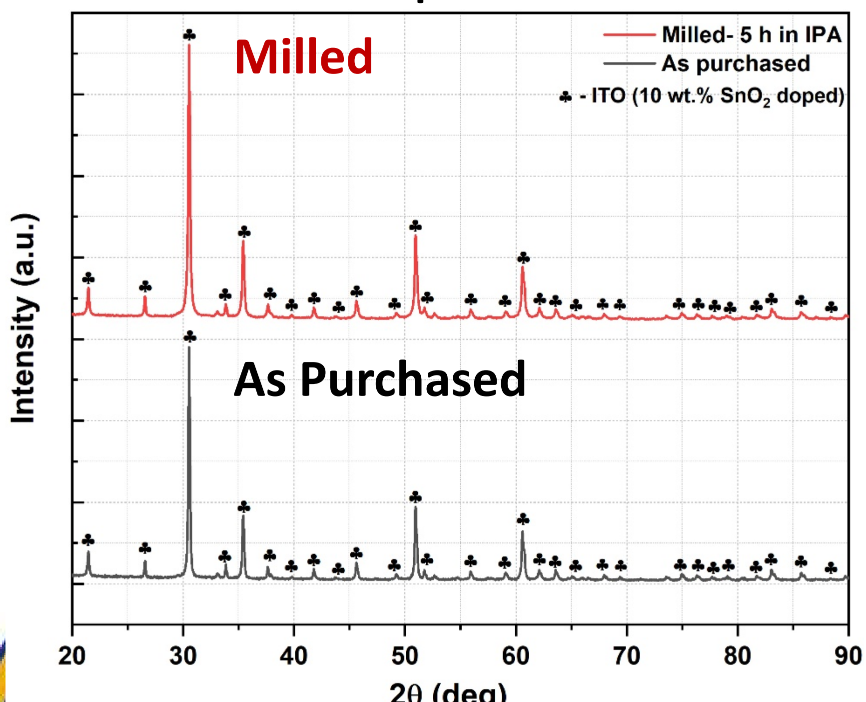


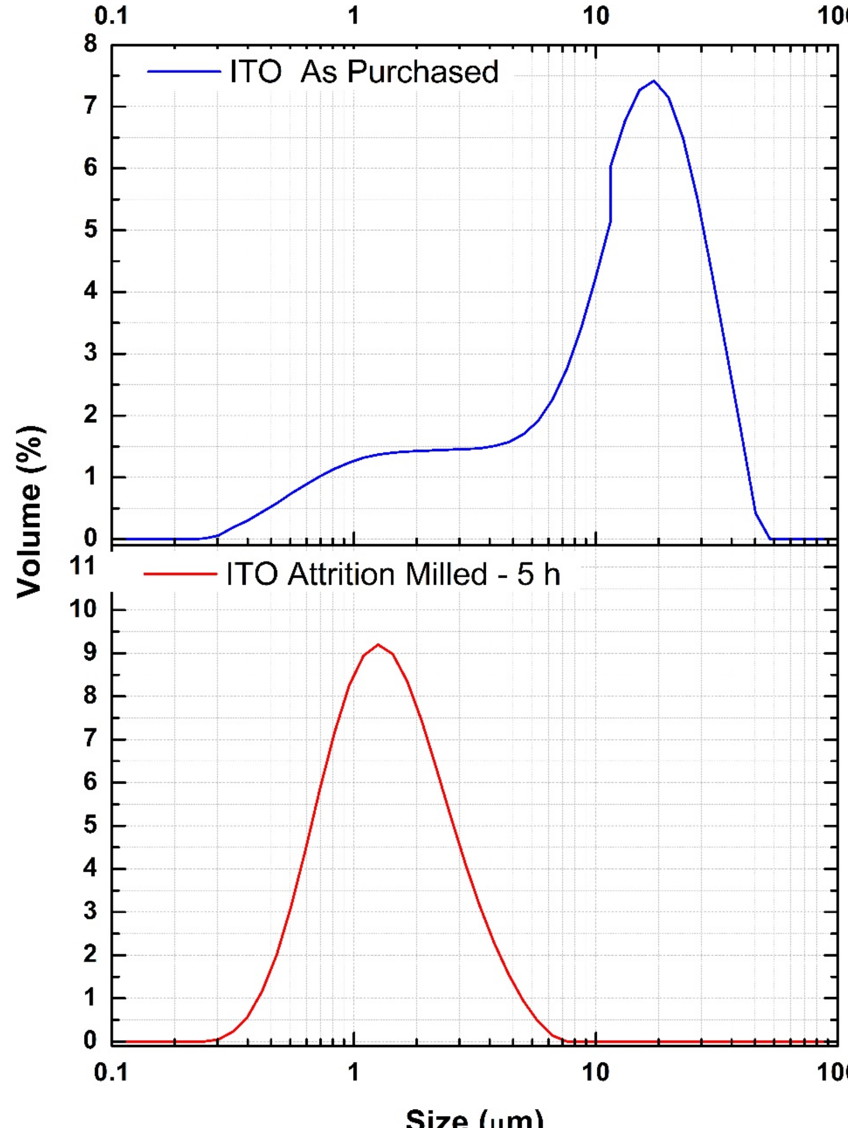


- **50-50 La_2NiO_4 – Al_2O_3 has a σ ~0.01 – 0.1 $S{\cdot}m^{-1}$ from 500 – 1100°C.**
- **60-40 La_2NiO_4 – Al_2O_3 has a σ ~0.01 – 1.05 $S{\cdot}m^{-1}$ from 500 – 1100°C.**
- **60-40 La_2NiO_4 – Al_2O_3 becomes more conductive at ~800°C.**

System 2a: Conductive oxide- Indium Tin Oxide (ITO)

- Stable up to 1250°C*.
- σ = 30 – 300 Scm^{-1} (RT – 1200°C)*.
- CTE~(6.2-8.1)x10^{-6} /°C.
- ITO: 10 wt% SnO_2 doping was used as purchased.
- Milled to achieve particle size <6 µm for sensor fabrication process.

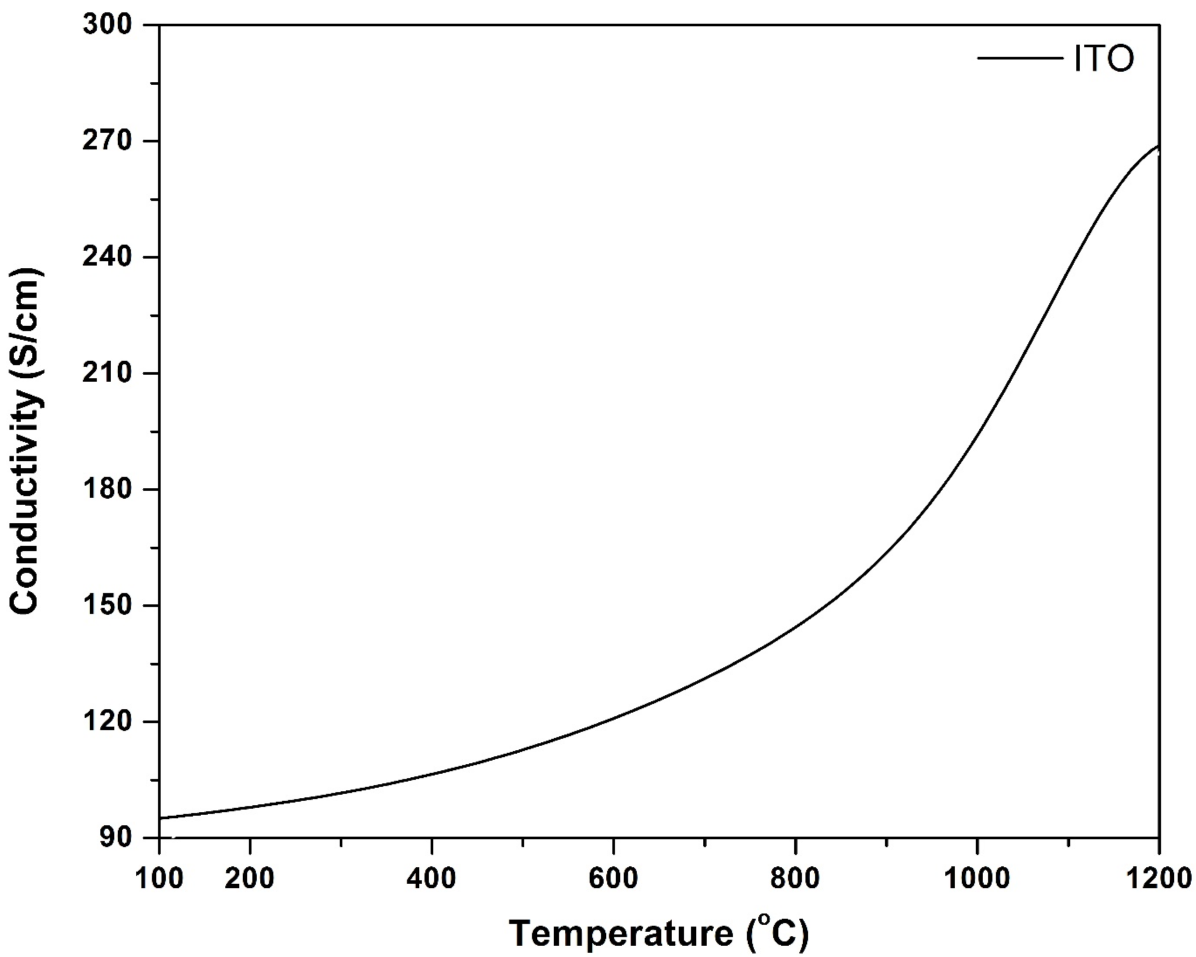




*Journal of Applied Physics **83, 2631** (1998), Sensors 2018,**18,958**

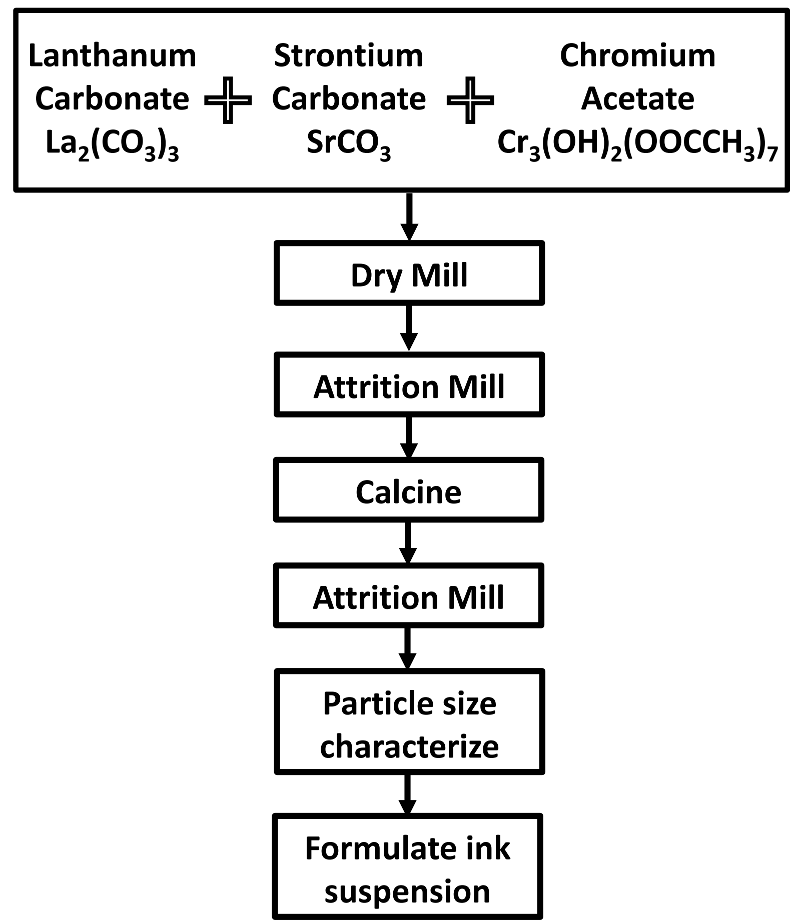
System 2a: Electrical Conductivity Indium Tin Oxide (ITO)

- Thick film conductivity (σ) with thickness ~35 µm was tested from 100 – 1200°C.
- σ = 93 – 270 $S \cdot cm^{-1}$ within the temperature range.



System 2b: Conductive oxide- Lanthanum Strontium Chromate (LSC)

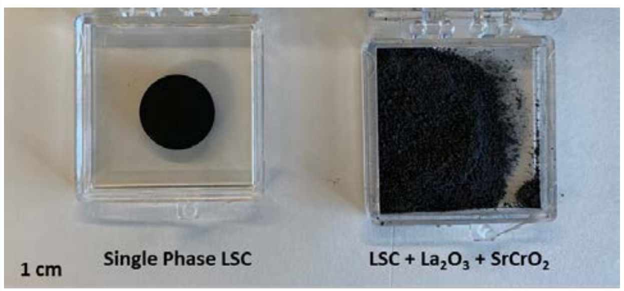
- Stable up to 1500°C*.
- σ = 10 – 200 S·cm^{-1} (RT – 1500°C)*.
- Compatible with refractory oxides (Al_2O_3, ZrO_2).

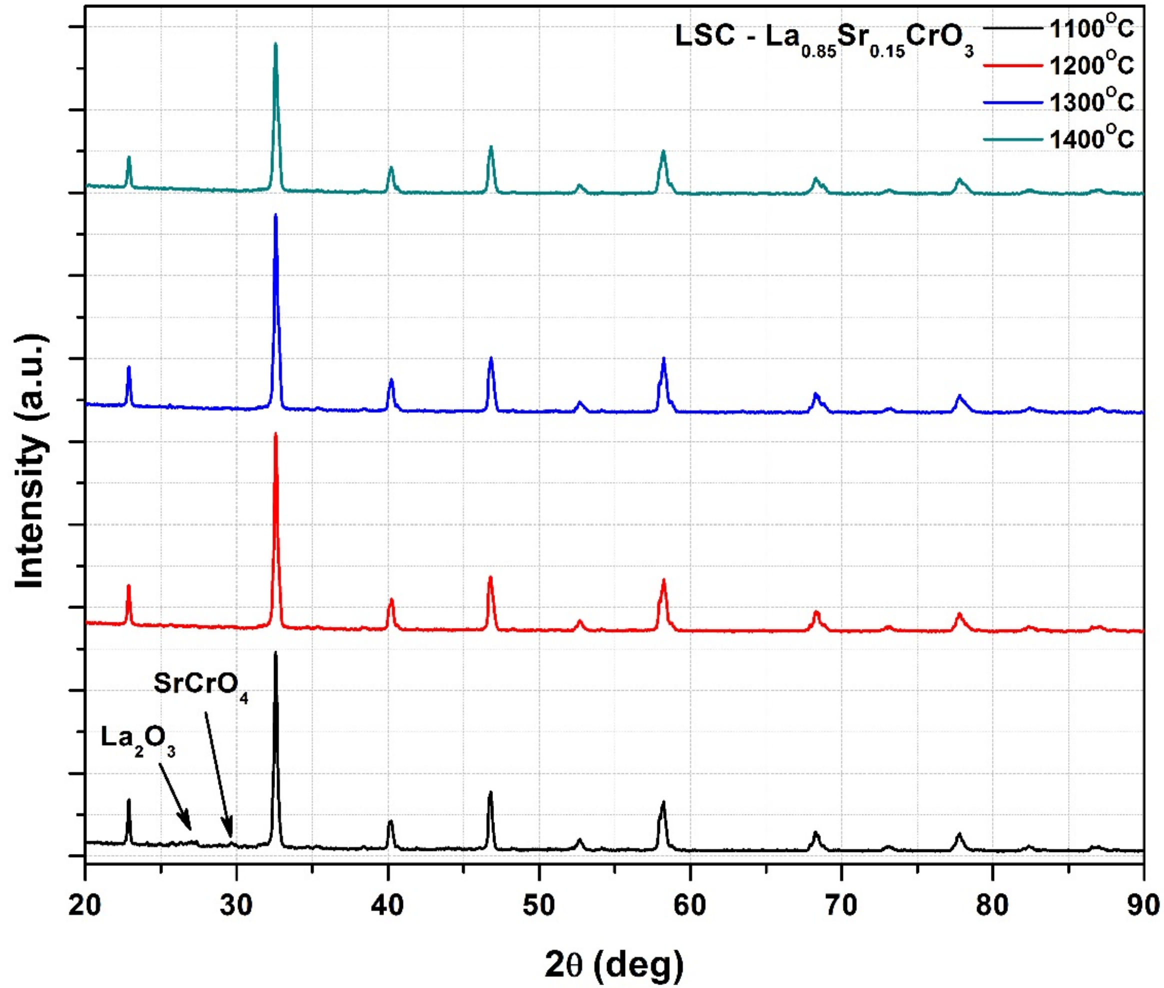


*D B Meadowcroft 1969, J. Phys. D: Appl. Phys. **2 1225**

System 2b: Conductive oxide- Lanthanum Strontium Chromate (LSC)

Pure $La_{0.85}Sr_{0.15}CrO_3$ should be obtained to avoid catastrophic hygroscopic behavior of residual La_2O_3.





Task 2.0 Summary:

- Synthesized and characterized composites of lanthanum nickelate, indium tin oxide and lanthanum strontium chromate.
- The phase stability, compatibility with the substrate and electrical conductivity was analyzed.
- Among the oxides, ITO and LSC showed good electrical conductivity for thermoelectric applications.

Task 2.0 Near-term Future Work:

- Synthesize:
  - Silicide-carbide systems with active and inactive (Al_2O_3, ZrO_2) fillers.
  - Other conductive oxide based composites.
- 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 electroceramic composites onto different surfaces.
- 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 method to transfer the pattern from an organic film to a ceramic surface and bond after thermal treatment.

Task 3.0 Approach:

Subtask 3.1 Direct-Writing 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 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.

Subtask 3.2 Micro-Casting Process Development:

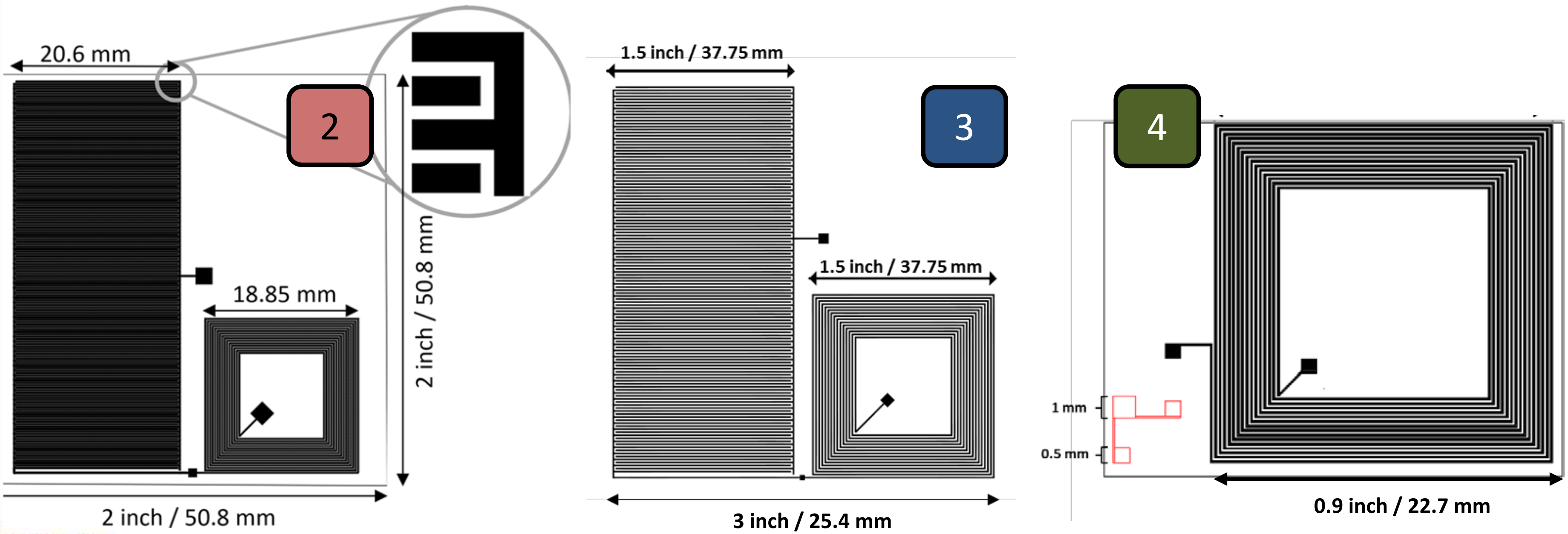
- ✓ Developed a micro-casting process based on photolithography to pattern reduced geometry sensor structures.
- ✓ Including methods to alter the wetting and drying characteristics of the deposited composite solutions.

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.

Various LC Architectures for the Sensor Fab

- Four architectures were developed using ANSYS/CAD drawing software based on the working frequency range.
- **Design 1:** 4-inch inductor/capacitor (IDC) pattern (used for low temperature initial assessment)
- **Design 2:** 2-inch inductor/capacitor (IDC) pattern (fabricated by micro-casting).
- **Design 3:** 3-inch inductor/capacitor (IDC) pattern (fabricated by direct writing).
- **Design 4:** 1-inch inductor/parallel plate capacitor (fabricated by micro-casting/



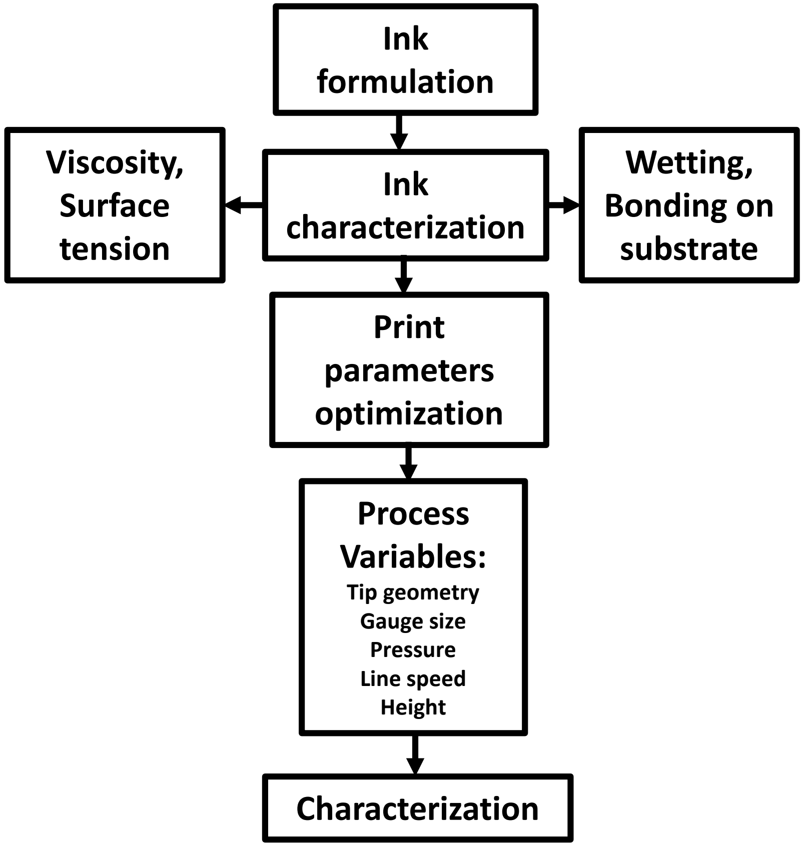
Initial Robo-Casting Evaluation:

Direct Writing:

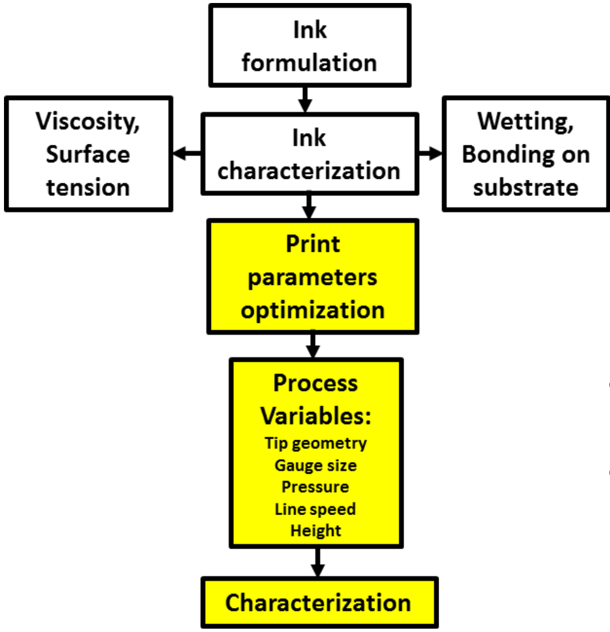
❖Developed inks within a permissible surface tension and viscosity level.

❖Direct-writing with Nordson EFD Performus VI robo-printer and Hyrel 3D.

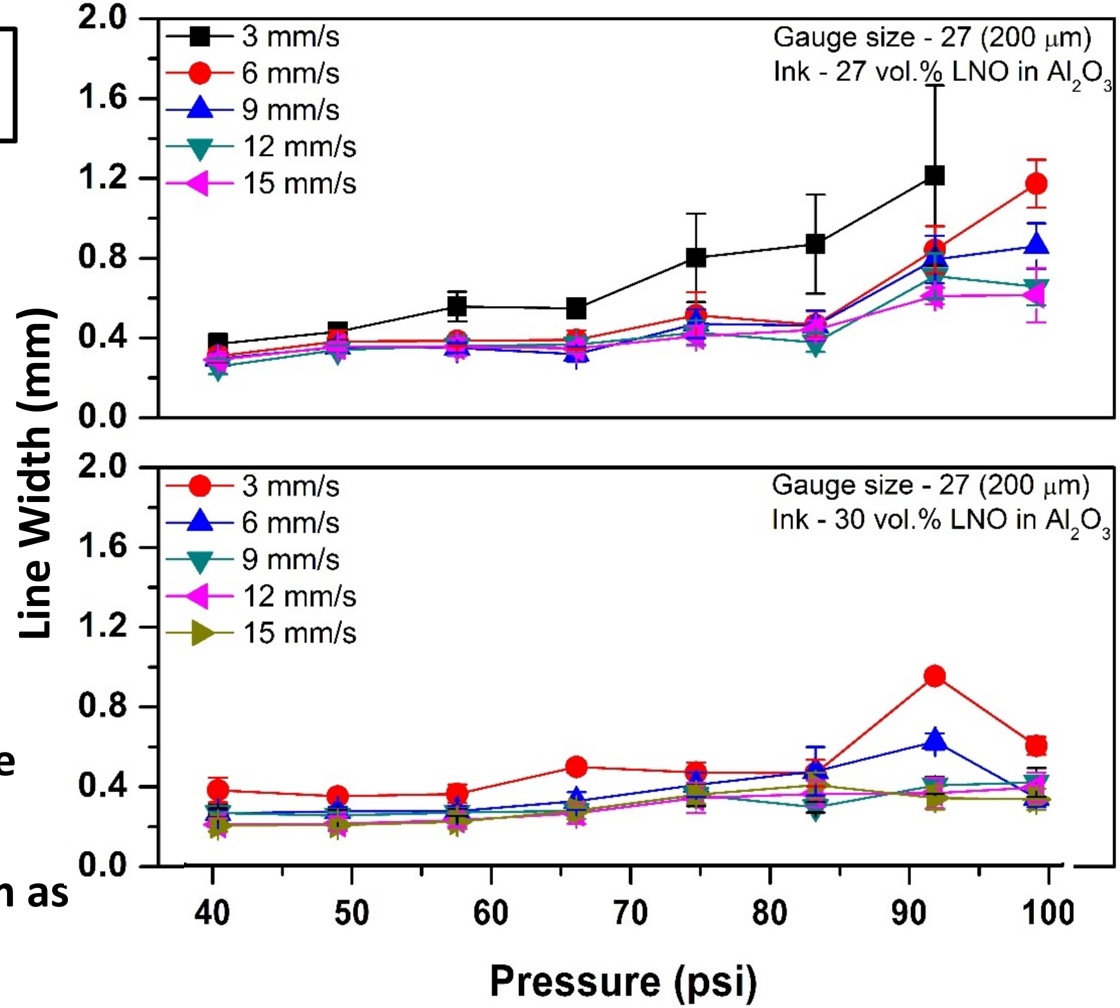
❖Initial evaluation of the ink and writing process was completed.



Initial Robo-Casting Evaluation:



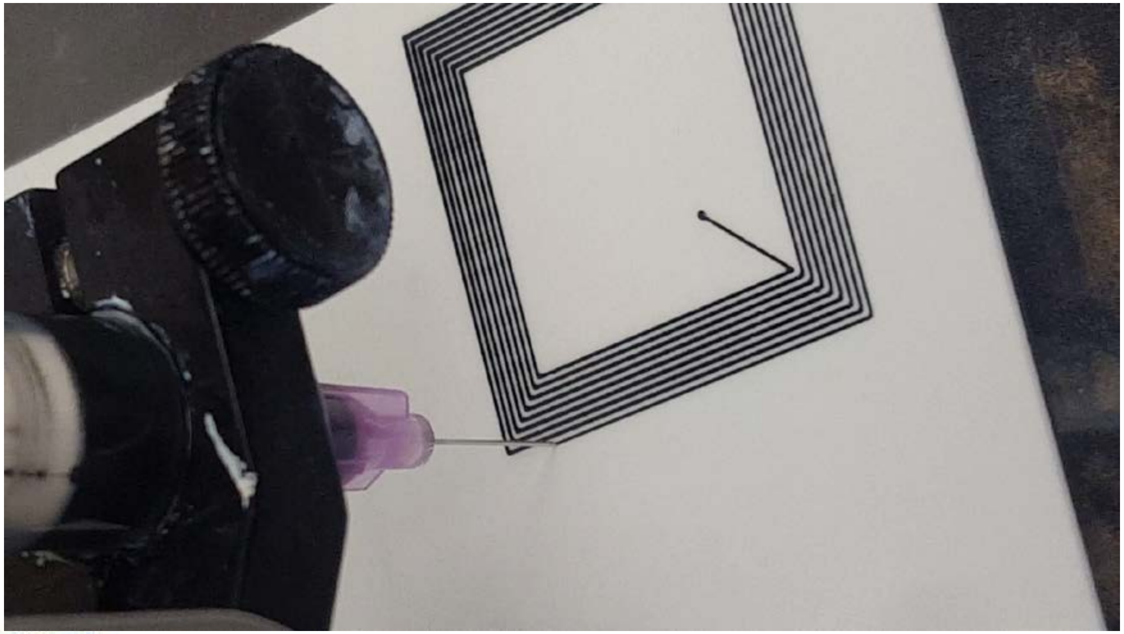
- **Line width depends on the pressure and print rate.**
- **Print rate determines the line width consistency.**
- **Line width as low as ~210 µm as achieved.**

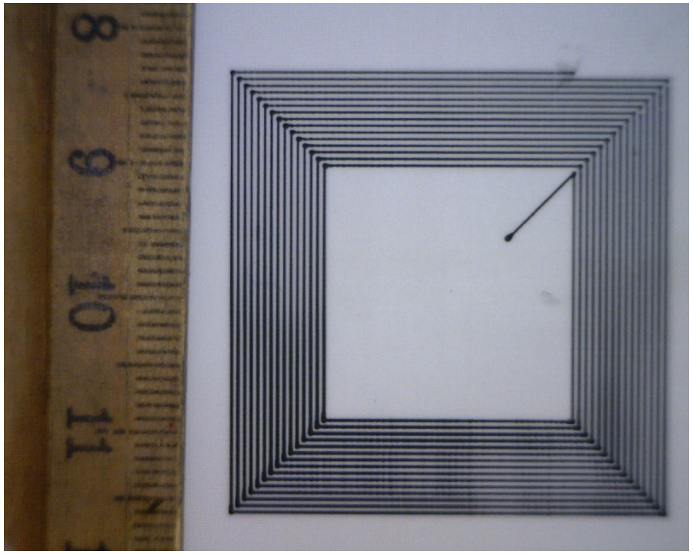


Demonstration of Direct Writing Process – Design 3

- **Parameters:**
  - Ink: 30 vol.%
  - Nozzle: 27 gauge (200 µm).
  - Print speed: 9 mm/s.
  - Line width : ~210 µm.
  - Line spacing: 350 µm.

- Larger form factor sensor (150 x 150 mm) was fabricated on alumina by direct writing process.





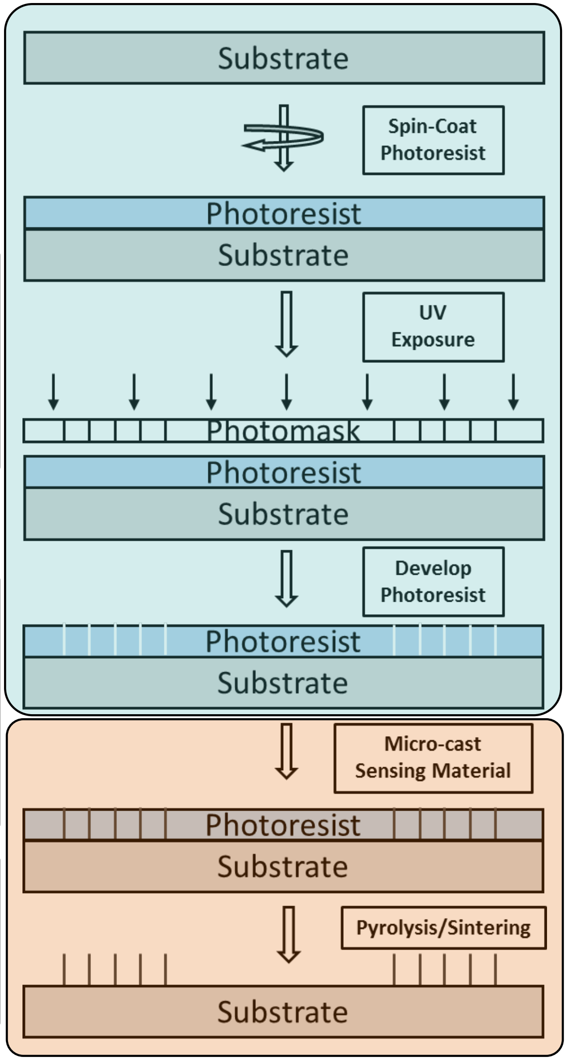
Micro-Casting Process: LNO-Al_2O_3 sensors – Design 2

Photolithography pattering and developing micro molds.

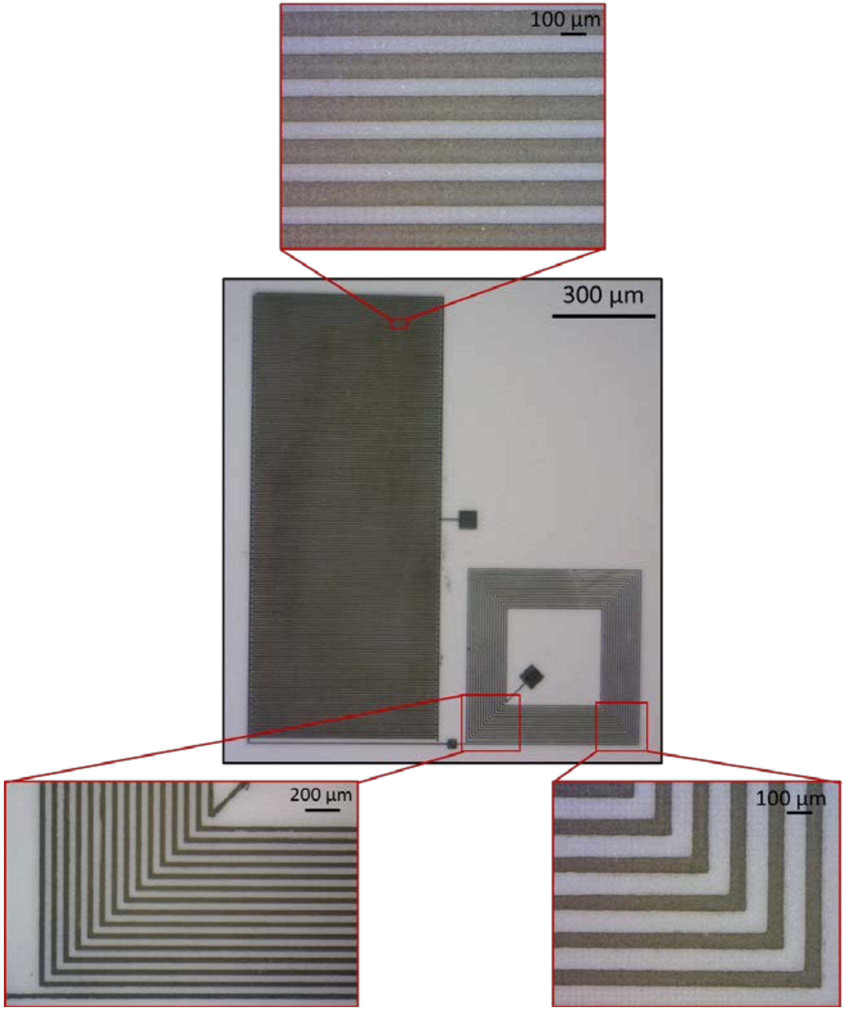
Ink formulation:

40 vol.% La_2NiO_4 – Al_2O_3 loading in ethyl cellulose/terpineol*

Casting and pyrolysis of the sensor.



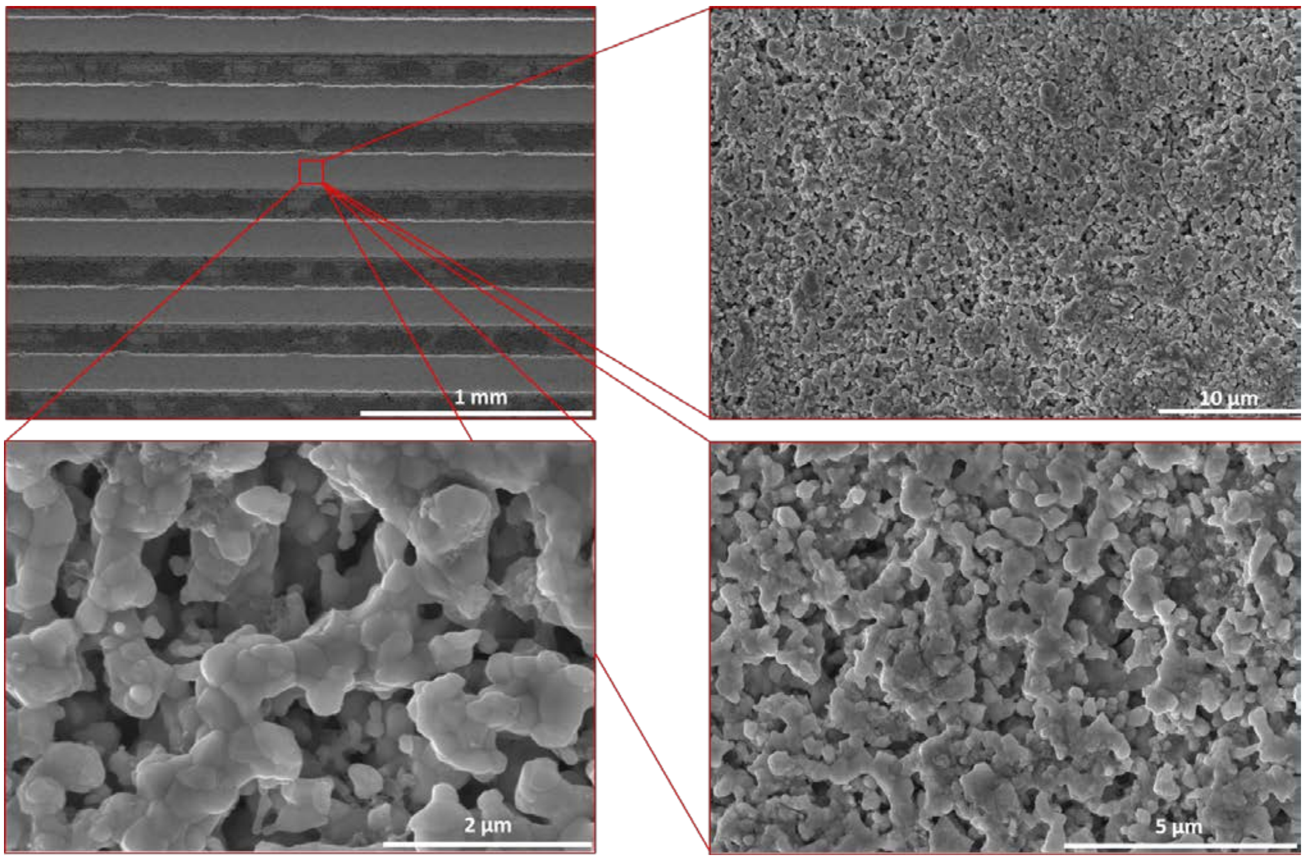
50.8 x 50.8 mm (2”) sensor on Al_2O_3 substrate.



*Johnson – Matthey Organic Ink Vehicle

Microstructure Analysis of 60-40 Sensor Composite

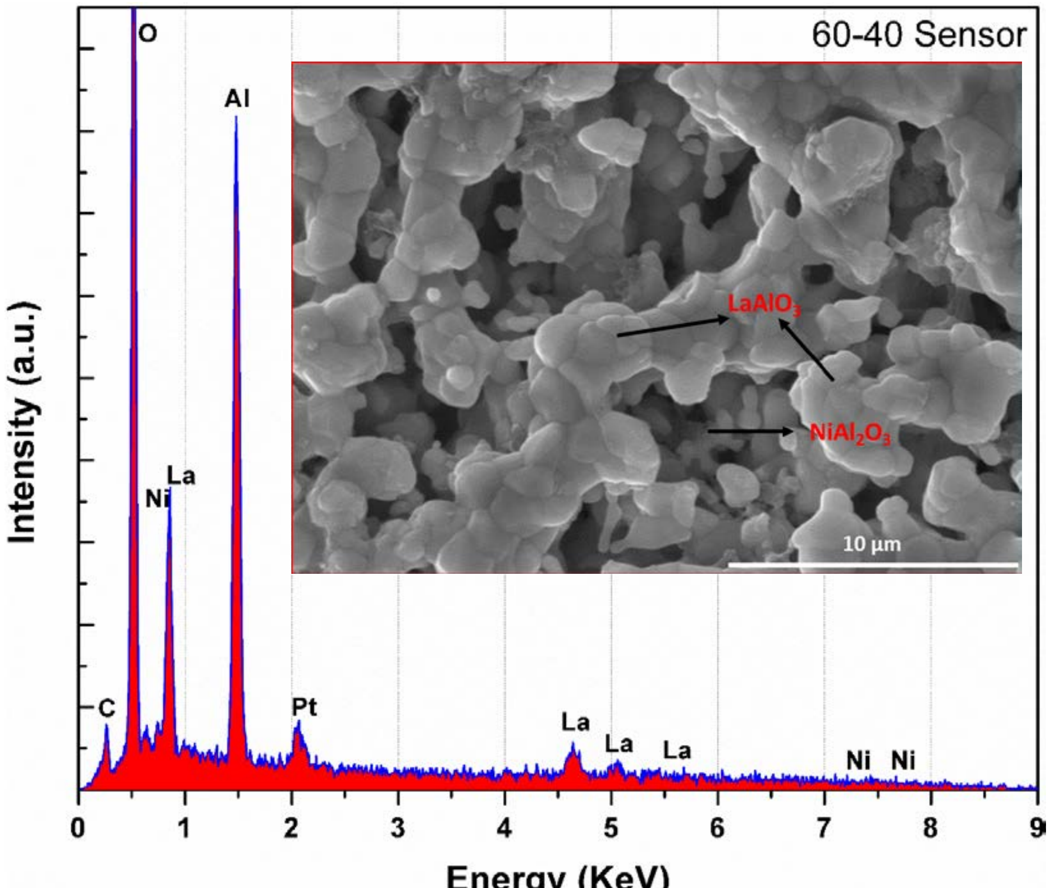
- SEM micrographs of inductor structure at different magnification.
- The micrograph shows the micro-casted sensor has good percolation.
- Average Grain Size: ~0.4 µm.



*Johnson – Matthey Organic Ink Vehicle

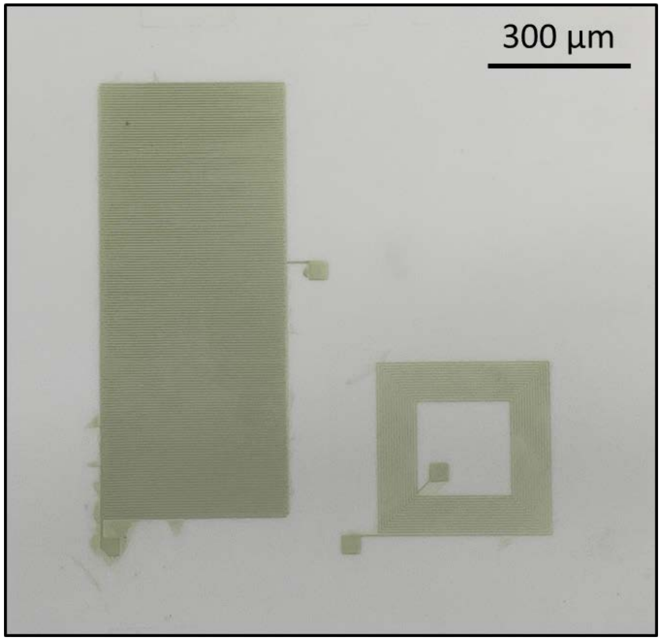
EDS Analysis of 60-40 LNO Sensor Composite

- **EDS micrograph shows the surface is predominantly covered by $LaAlO_3$ phase.**
- **The $NiAl_2O_4$ phase has smaller grains and percolated under the $LaAlO_3$ phase.**
- **Pt arises from the conductive metal layer coating to avoid charging effect.**

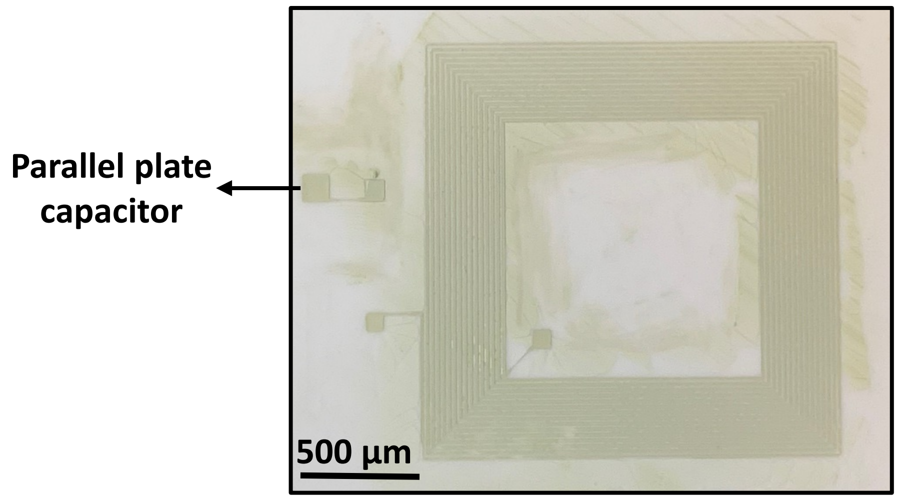


Micro-Casting Process: ITO Sensors

50.8 x 50.8 mm (2”) sensor (design -2) on Al_2O_3 substrate.



25 x 25 mm (1”) sensor (design -4) on Al_2O_3 substrate.



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.
- Initial direct analysis of the particle dispersed in polymer ink was evaluated to fabricate LNO-Al_2O_3 sensors (design 2).
- Microstructural stability of the LNO-Al_2O_3 sensors were evaluated for isothermal hold up to 96 h at 1050^{o}C showing no change in the grain size.

Task 3.0 Near-term Future Work:

- Robo-cast preceramic ink precursors (ITO and LSC) onto oxide substrates and optimize firing temperature for optimum density.
- Achieve print resolution ~150 – 200 um to print next generation LC circuits.
- Translate the technology and print LSC on alumina substrate to carry out wireless characterization up to 1500^{o}C.

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

Task 4.0 Objectives:

- Design and model passive wireless sensors and interrogator antennas as RLC circuits
- Fabricate and test the sensor up to 100^{o}C
- To extend sensor performance up to 1500^{o}C
- Advanced materials/writing processes => existing sensing strategies don't work!
- ***Innovation:*** *advanced materials, advanced processes,* ***robust and adaptive signal processing***

Task 4.0 Approach:

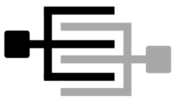
- Design RLC circuits via simulation for various material properties
- Develop a robust and adaptive signal processing approach to measure temperature wirelessly. *Ink/substrate material characteristics may not be know precisely or may change with heating/cooling cycles **=> tracking the resonant frequency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

Task 4.0 Current Status:

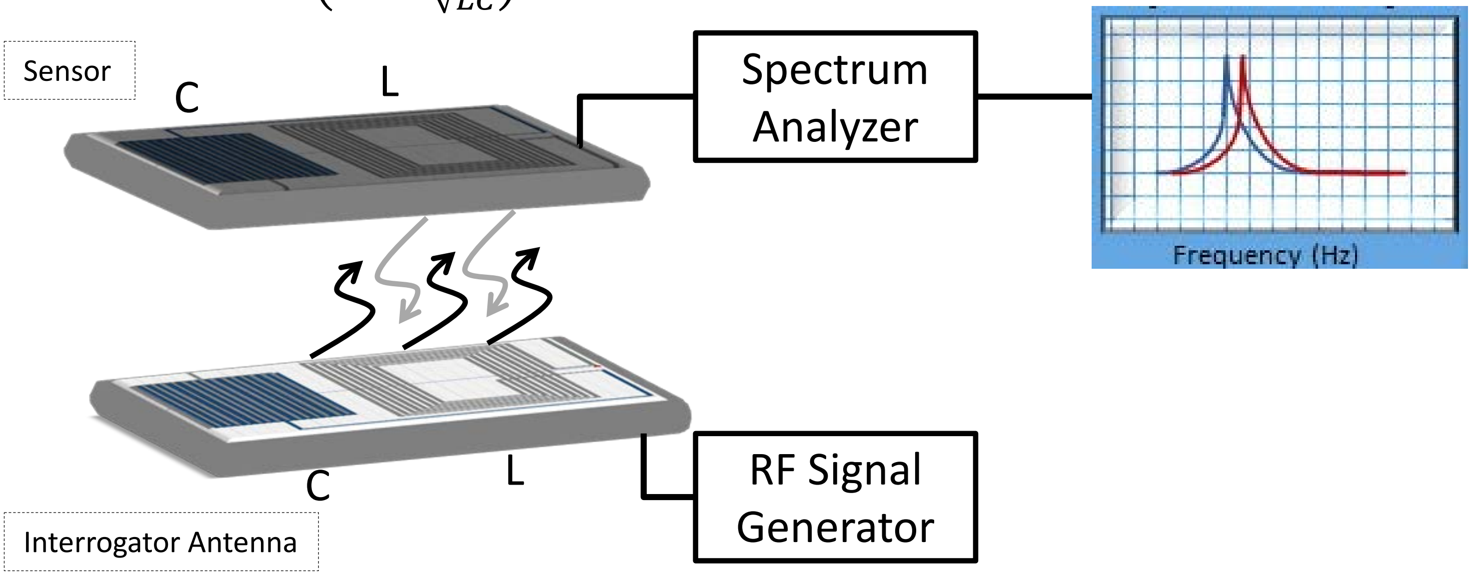
- ✓ Completed wireless characterization of LNO-Al_2O_3 (design-2) sensors from 500 – 1000°C.
- ✓ New algorithms were developed to replace conventional signal processing techniques for improves data analysis.
- ✓ Begun high-temperature characterization with pure oxide based electro-ceramic material systems: ITO and LSC.
- ✓ Reduced sensor size: 4 inch -> 2 inch -> 1 inch

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

- An 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) $\longrightarrow$ 

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



How is our problem different?

- Lots of Unknowns:
  - Material electrical properties
  - Ink electrical properties
  - interconnect (wires/pads) effects
  - Temperature (what we're trying to measure)

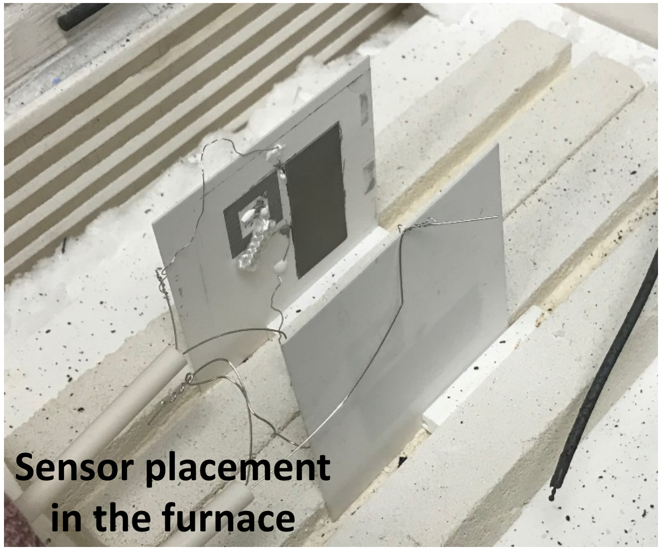
- **Material/ink/interconnect properties change with time and with heating/cooling cycles**

- It's easy to design for a particular resonant frequency, *but that doesn't mean it will actually resonate*. **We need a novel robust adaptive approach!**

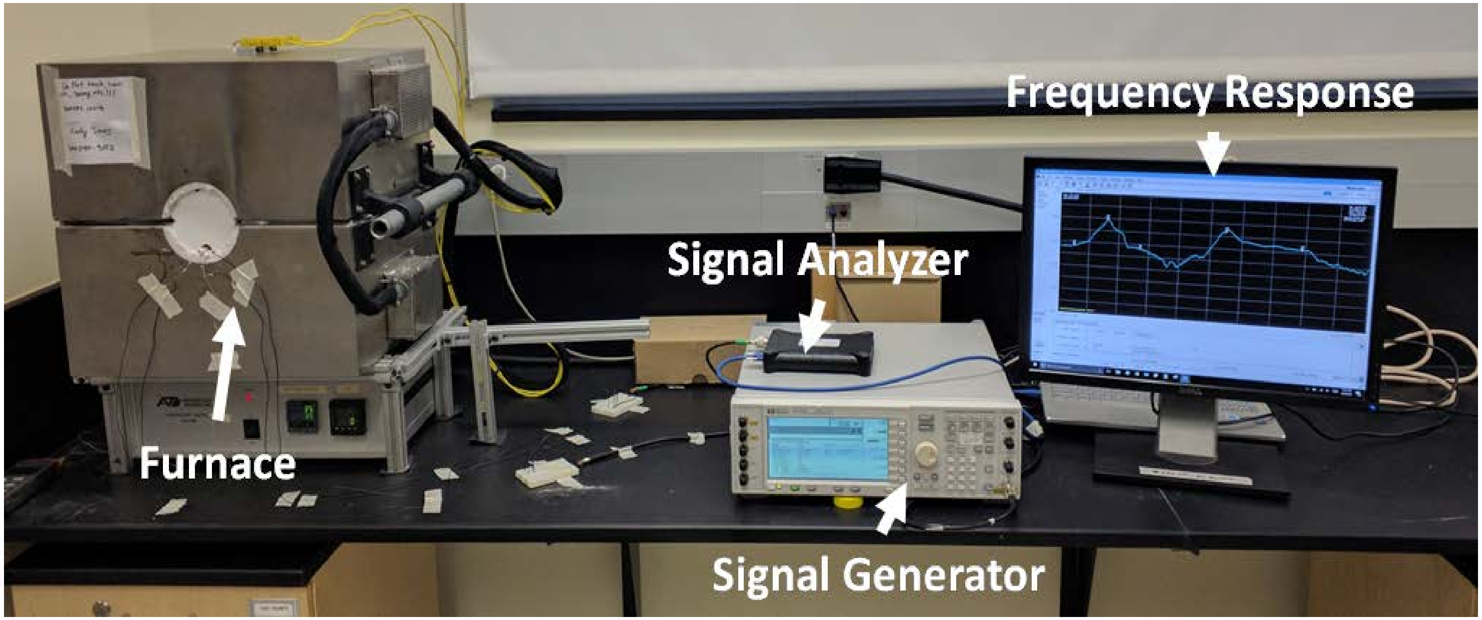
Our Signal Processing Approach

- **Don’t rely on a single resonant frequency.**
  - transmit across a wide spectrum (sweep or impulse) around the resonant frequency
  - store wideband *signatures* at known temperatures of interest, say every 25C (create a database)
  - sweep at unknown temperatures, compare signal to database of signals at known temperatures
- Creates a signal matching problem instead of a tracking problem => **more degrees of freedom**
- Leverage research on signal matching for biometrics, digital communication, RADAR, SONAR, radio astronomy, …
- Our scheme is **adaptive**: update signatures as needed
- Our initial scheme: correlation and maximum absolute error

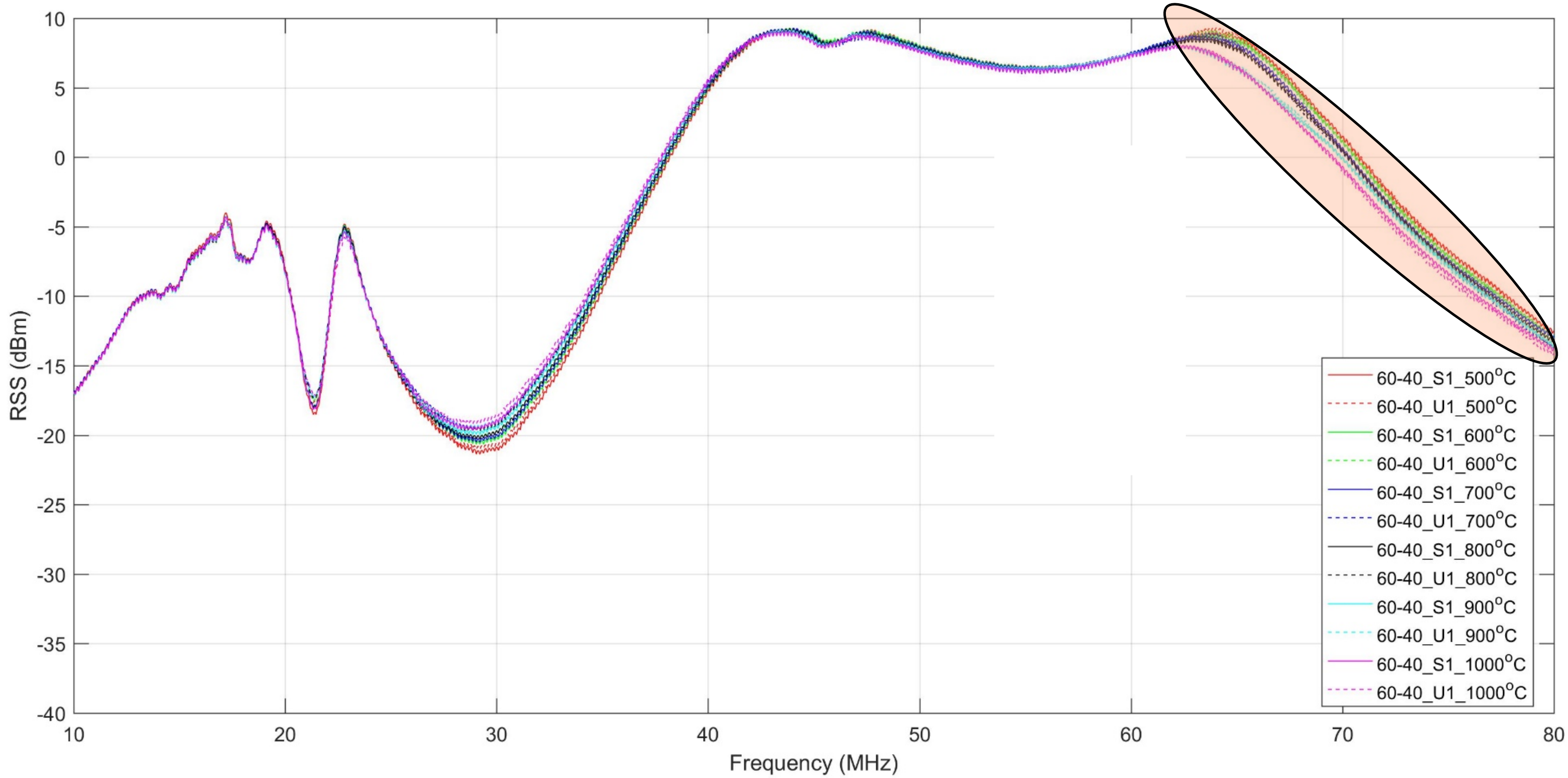
Wireless Characterization Setup



- Two sensors were used to characterize the wireless signal response.
- One is connected to signal generator and the other to signal analyzer.
- Distance between the sensors ~10 cm.

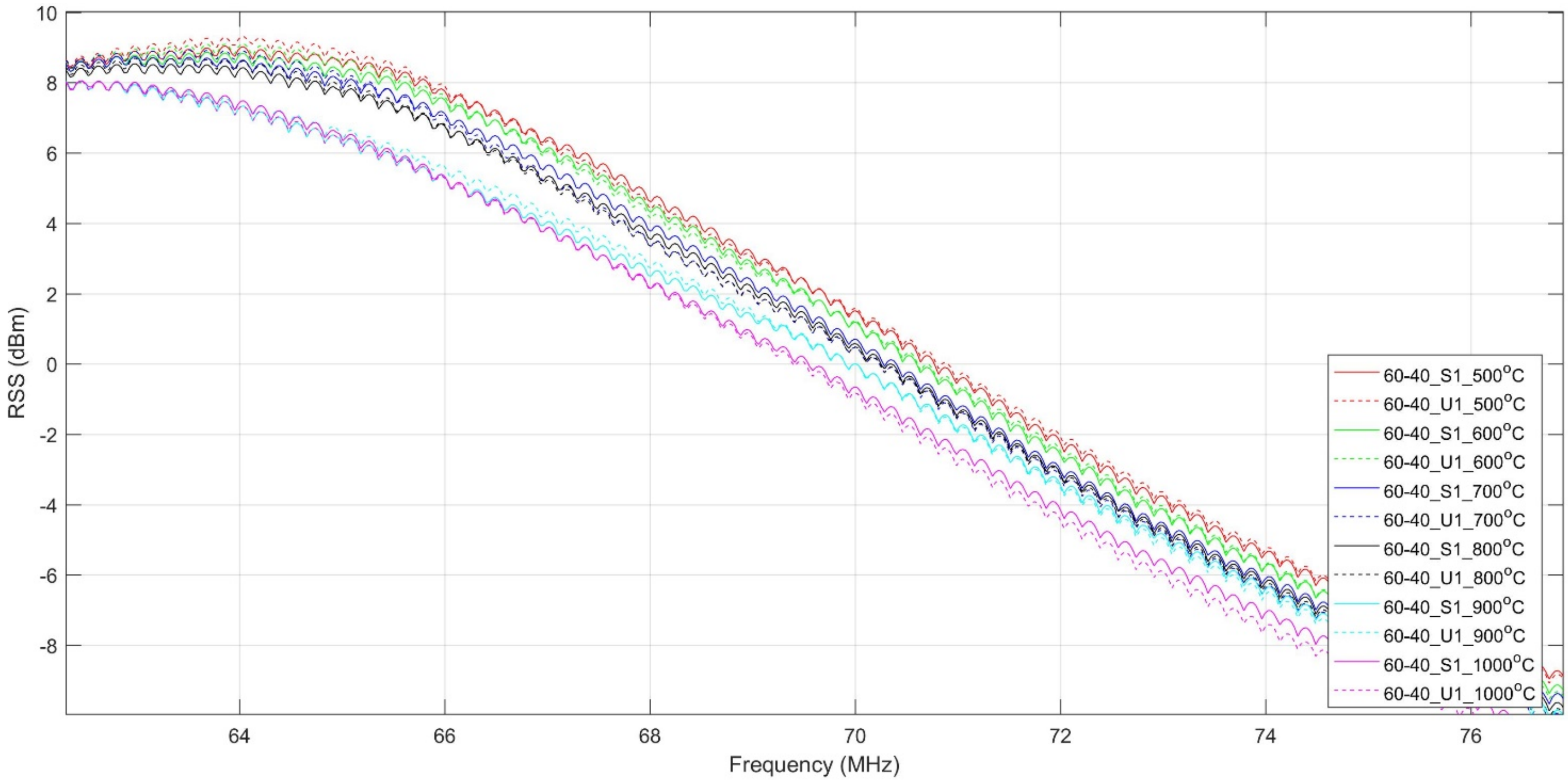


Example Sensor Sweep (Sensor Design 2)



- **6 signatures and 6 unknowns over a wide spectrum (500 – 1000°C).**
- **Temperature signatures are unique and distinguishable with a sensitivity of 0.2 kHz/°C.**

Where do the curves differ most (optimize the window)



- **6 signatures and 6 unknowns over a wide spectrum (500 – 1000^{o}C).**
- **Temperature signatures are unique and distinguishable with a sensitivity of 0.2 kHz/^{o}C.**

Frequency Analysis & Signal Processing

✔ - Match

✗ - Not a match

- **Match (cross-correlation) using the entire spectrum:**

500ºC- ✔ ✔; 600ºC - ✔ ✔; 700ºC- ✗ ✗; 800ºC- ✔ ✔; 900ºC- ✔ ✔; 1000ºC- ✔ ✔

Performance is not good enough!

- **Use only the "best part" of the spectrum (53MHz to 69MHz):**

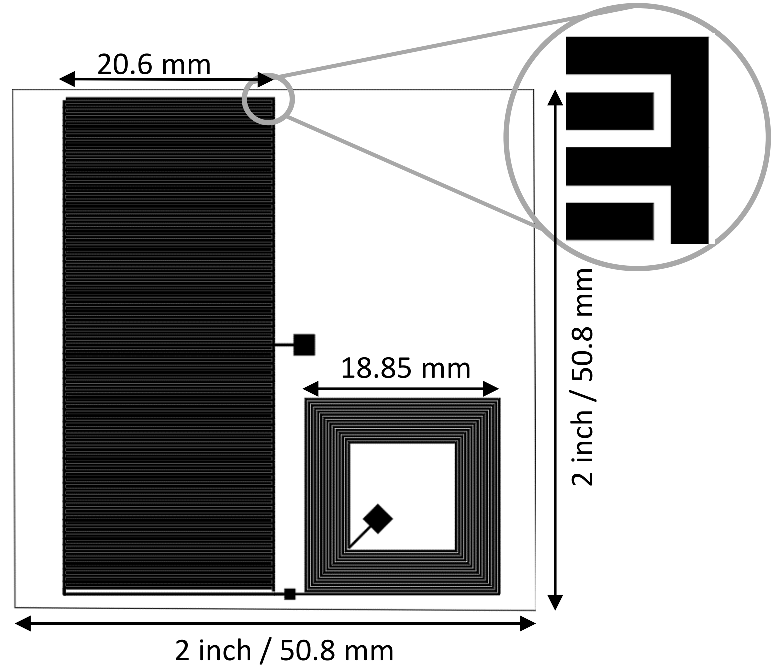
500ºC- ✔ ✔; 600ºC - ✔ ✔; 700ºC- ✔ ✔; 800ºC- ✔ ✔; 900ºC- ✔ ✔; 1000ºC- ✔ ✔

Good performance!

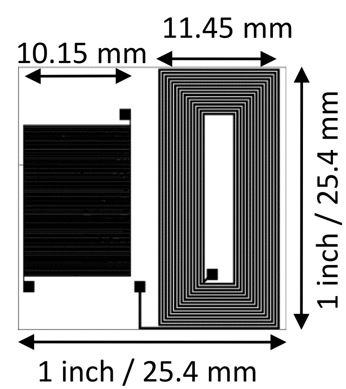
Signature	10.0000	10.0292	10.0583	10.0875	10.1167	10.1458	10.1750	10.2042	10.2333	10.2625
Unknown	10.0000	10.0292	10.0583	10.0875	10.1167	10.1458	10.1750	10.2042	10.2333	10.2625

Reducing the Form-Factor

- 2 inch sensor
  - IDC
    - Number of fingers : 80
    - Width of each finger : 0.2 mm
    - Gap between fingers : 0.1 mm
  - Inductor
    - Number of turns : 15
    - Width : 0.15 mm
    - Gap : 0.15 mm



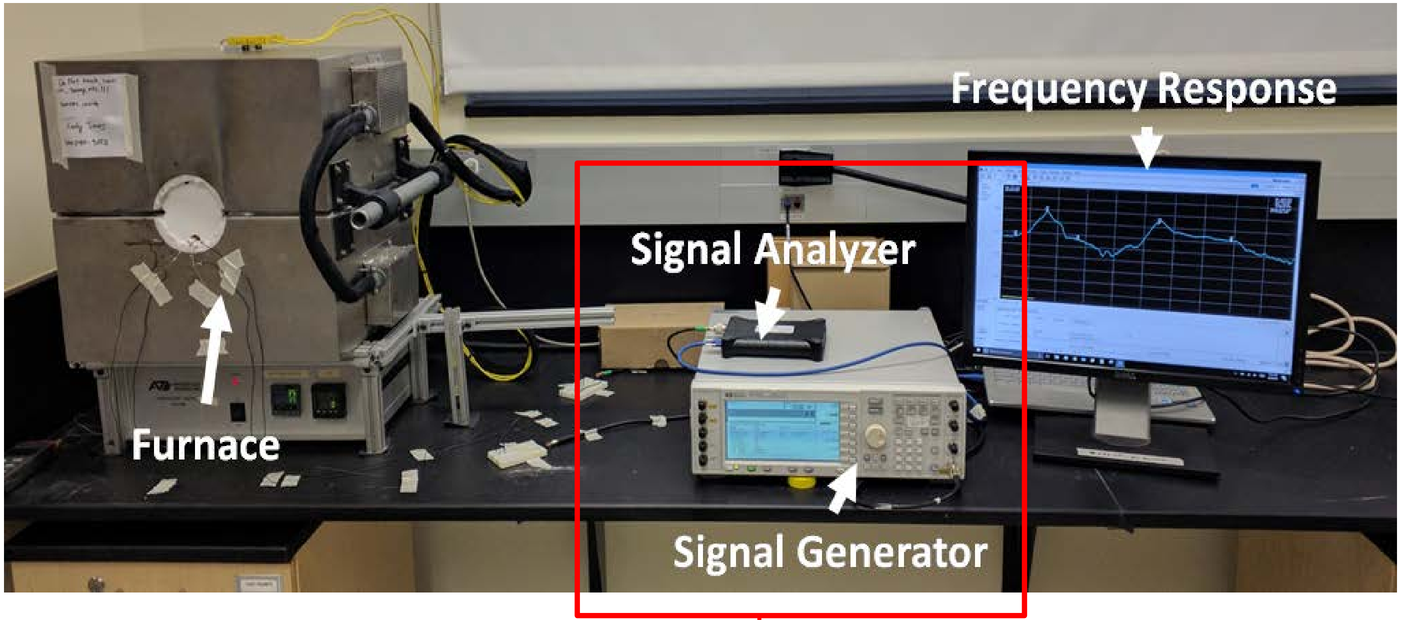
- 1 inch sensor
  - IDC
    - Number of fingers : 96
    - Width of each finger : 0.05 mm
    - Gap between fingers : 0.025 mm
  - Inductor
    - Number of turns : 15
    - Width : 0.15 mm
    - Gap : 0.15 mm



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), Matlab, Labview

USRP Usage in current setup



Task 4.0 Summary:

- Completed preliminary 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
  - Built smaller form-factor 3D models and simulations of sensor designs

Task 4.0 Near-term Future Work:

- Work with newer sensor designs to investigate the high temperature performance of the sensors.
- Build a low-cost USRP powered wireless sensing platform

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