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Development of Anisotropic Thermal Management Materials

PRESENTED BY

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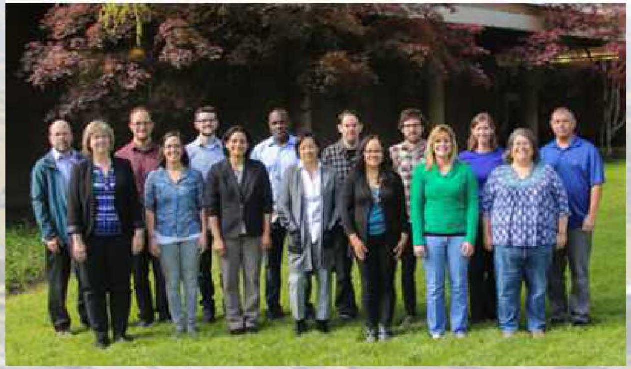
Chemistry, Combustion & Materials Science

Materials Research

Materials Chemistry Department



The Materials Chemistry Department provides creative solutions to materials problems impacting national security.

Polymer Lab

Synthesis Lab

MAC Lab

Plating Lab

Thermal-Elec Lab

Thermal-Physical & Electrical Lab

- Materials characterization of a wide range of materials (from foams to metals),in different states and morphologies (liquid, thin coatings, composites…) and under different accelerated environments (temperature, humidity).

Materials:
polymers, metals, ceramics, composites, dielectric, thermoelectric, anisotropic materials…

Properties:
Thermal conductivity, diffusivity, heat capacity, latent heat, dielectric constant, capacitance, resistivity…

Applications:
R&D of new materials
Qualification under different environments
Lifetime and Aging studies

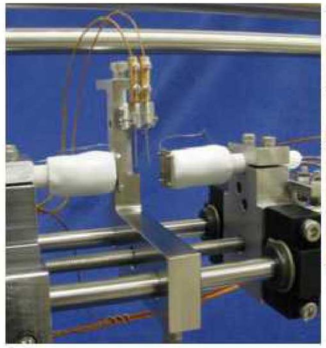
- Thermal, physical and electrical testing using state of the art and custom methods for accurate and realistic materials characterization.

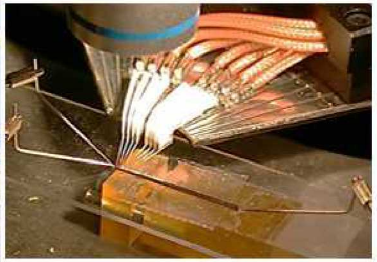
Thermophysical System

TGA-DSC

High and ultra low resistivity setup

Custom designs

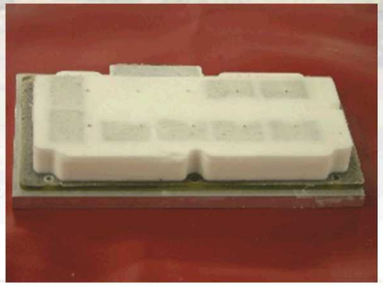


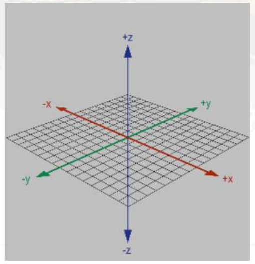


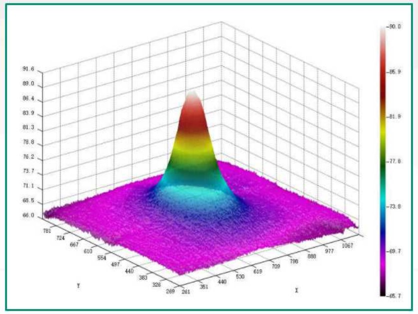
Micro-probes

Development for Engineered Anisotropic Thermal Barriers

Karla Reyes (PI), Jacob Mayer and Austin Acosta

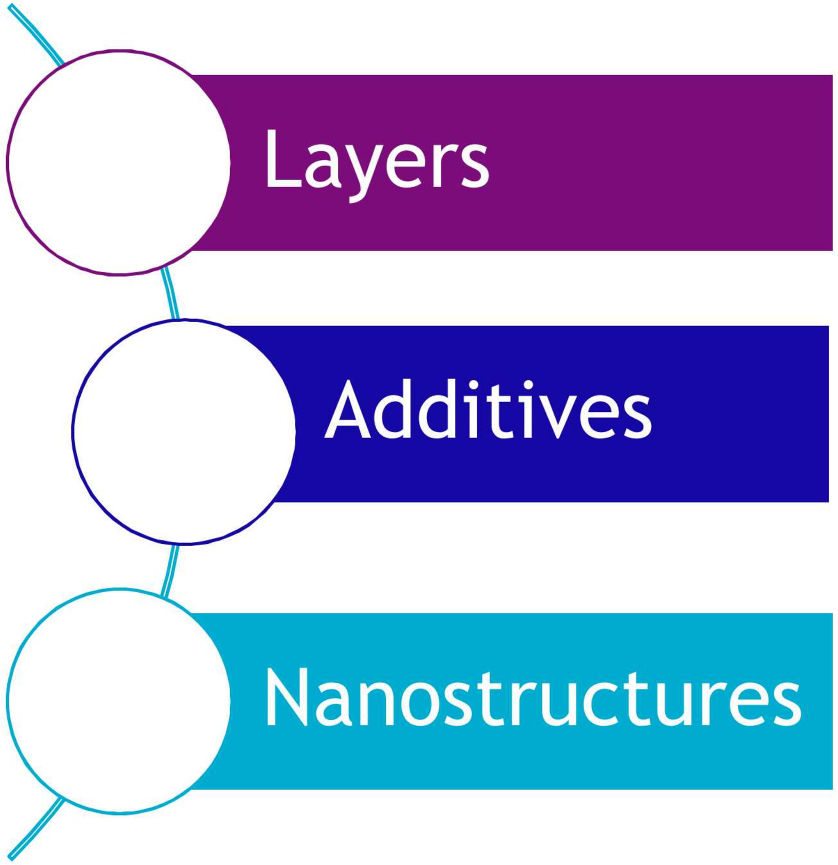






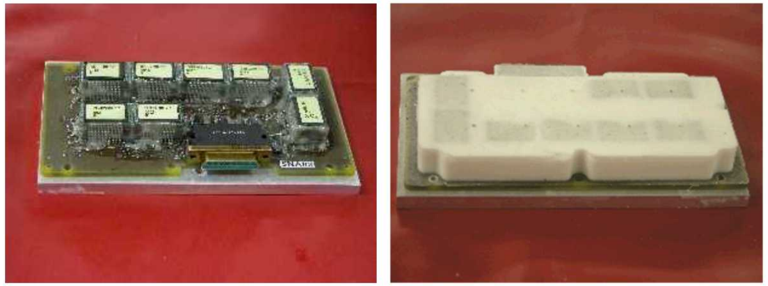
Outline

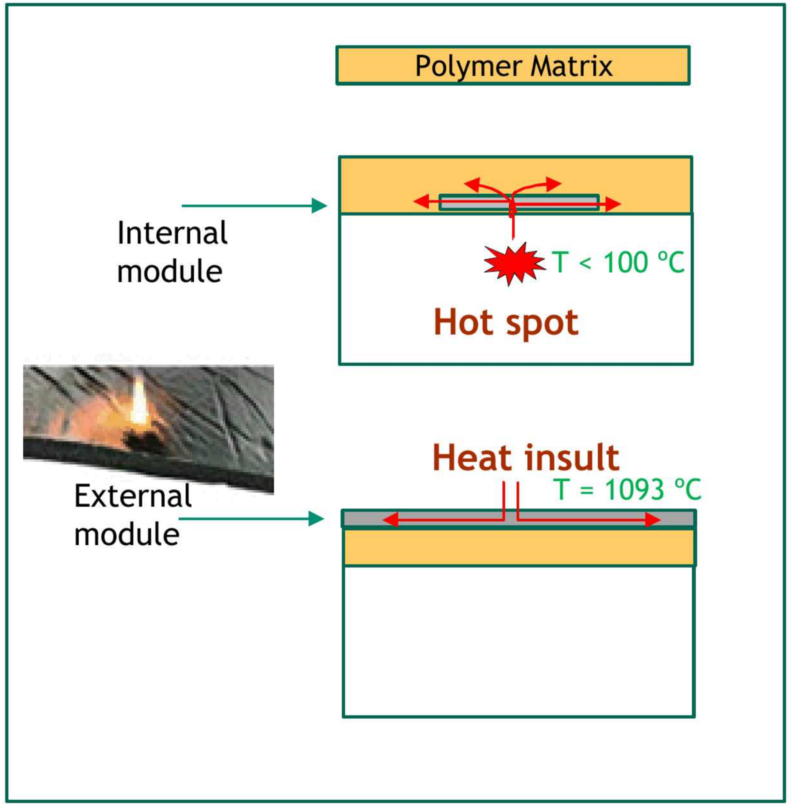
- Thermal management needs
- Engineered anisotropic thermal barriers
- Experimental and modeling gaps
- Anisotropic thermal properties measurements
- Predictions and modeling efforts
- Future directions



Thermal Management Needs

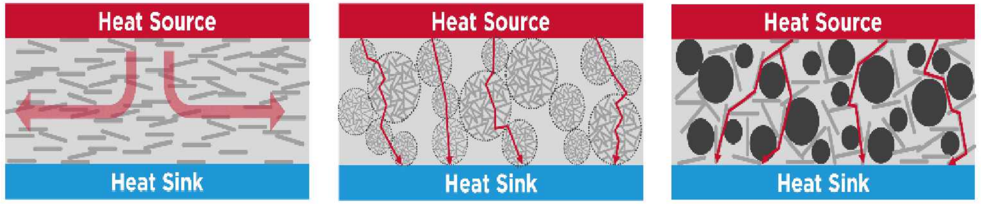
- Encapsulation materials (such as foams) for mechanical protection deal with problems arising from heat dissipation, thermal stresses, and warping due to high temperatures.
    - internal overheating
    - external heat insults
- How anisotropic thermal materials can help?
    - Anisotropic thermal barriers can be engineered to optimize the heat transfer in-plane and through-plane directions.
        - ✓ Addition of internal high conductive layers to create fast heat dissipation pathways from electronics to heat sinks.
        - ✓ Addition of external layers to protect thermal insulation from high temperature insults (such as fires).





How to engineer anisotropy in thermal barriers?

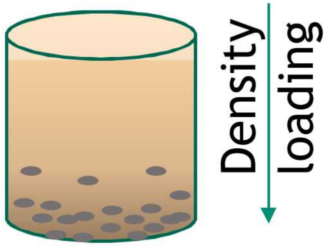
- Layers
- Additives
- Nanostructures

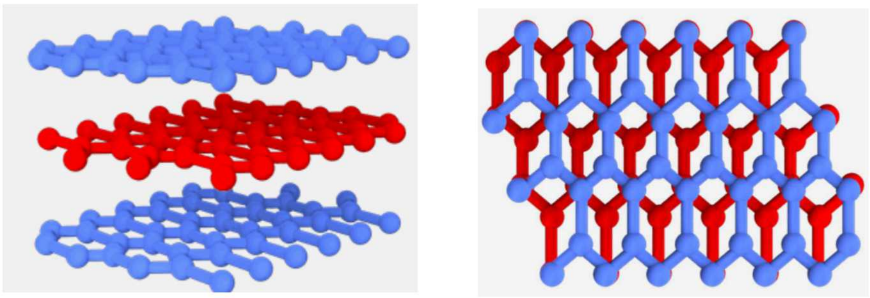


Platelets alone tend to align in the polymer flow direction, creating thermal pathways that are more effective for heat spreading.

Using hBN agglomerates helps to randomly orient the platelets, creating more isotropic thermal conductivity.

Combining BN platelets with non-acicular particles will disrupt alignment, improving through-plane thermal conductivity.





graphite

Materials Characterization Capabilities

➢Thermal Characterization Methodology

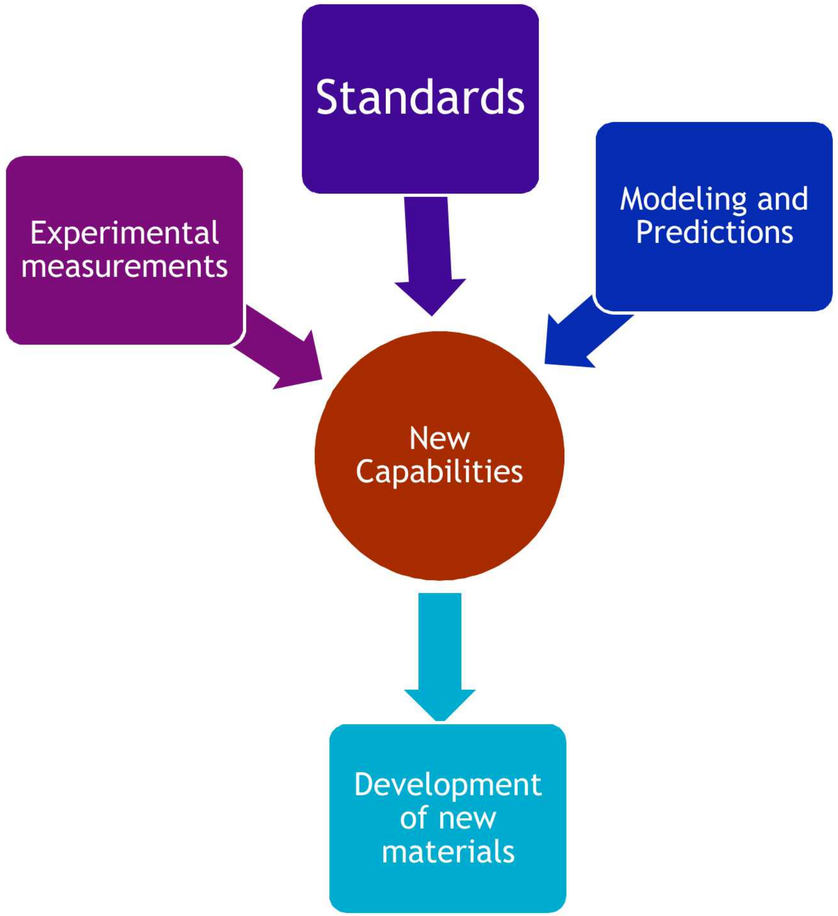
There are only three absolute methods of measuring thermal conductivity.

(1) Guarded Hot Plate – This method is only good for measuring low thermal conductivity materials.

(2) Laser Flash Apparatus – A well-known method for testing medium to high thermal diffusivity, but then the density and specific heat are used to calculate the thermal conductivity. However, the density and specific heat of composite or layered materials can be very challenging to measure, which makes this technique unreliable for anisotropic materials.

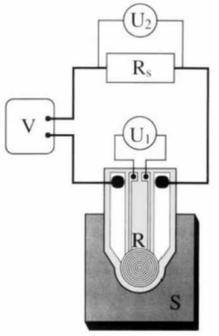
(3) Hot Disk Transient Plane Source (TPS) – A versatile method for testing low to high thermal conductivity materials, where the thermal conductivity is directly (or independently) measured.

- ➢Traditional methodology is not designed to measure anisotropic materials.
- ➢ New experimental capabilities coupled with analytical models to understand multi-materials thermal properties are urgently needed for accurate measurements and predictions of complex thermal barrier materials.

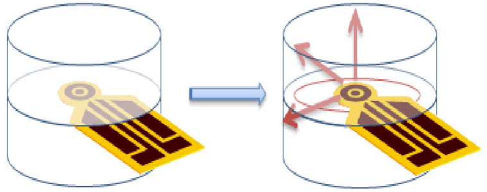


Hot Disk Transient Plane Source (TPS)

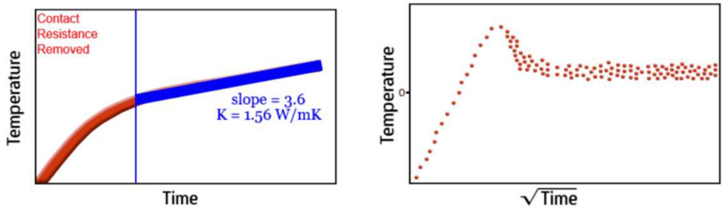
- A Hot Disk Sensor consists of an electrically conducting pattern on a thin sheet of Nickel. The sensor is fitted between two pieces of the sample, each one with a plane surface facing the sensor. The sensor applies an electrical current, high enough to increase the temperature of the samples up to several degrees. At the same time, the sensor records the resistance (temperature) increase as a function of time.
- The heat generated dissipates through the sample on either side at a rate dependent on the thermal transport characteristics of the material. By recording the temperature versus time response in the sensor, thermal conductivity can accurately be calculated.
- Anisotropic module can be used for homogeneous anisotropic materials assuming that the thermal properties in x and y directions are the same and different (but constant) in z directions.



Pictures of thermal property system and oven used in this work. (photos modified from Thermtest, Inc. website)



Scheme of how the Hot disk sensor is sandwiched in between two identical samples to perform the measurements.

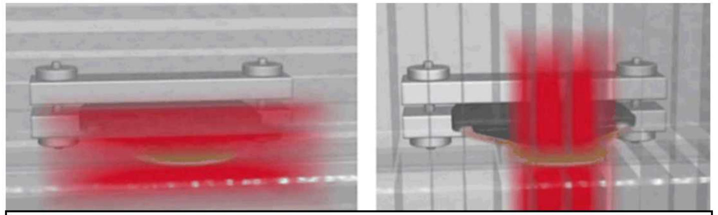


$$\Delta T_{ave}(\tau) + \Delta T_i = \frac{1}{\alpha} \cdot \left(\frac{R(t)}{R_0} - 1 \right)$$

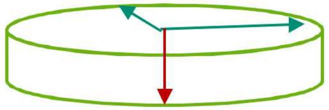
$$\Delta T(t) = \frac{P}{\pi^{3/2} \cdot r \cdot k} D(t) \qquad \tau = \sqrt{\frac{t}{\theta}} \text{ and } \theta = \frac{r^2}{\alpha_r}$$

Where
P = power
r = sensor radius
k = thermal conductivity
α = thermal diffusivity
ρc_p = volumetric specific heat

r = Radial a = Axial



TPS technique for anisotropic measurements. But only for homogeneous materials, no multilayers systems



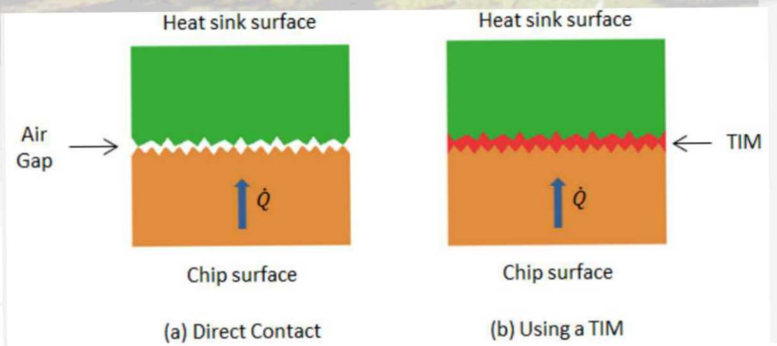
$$k = (k_r k_a)^{1/2}$$

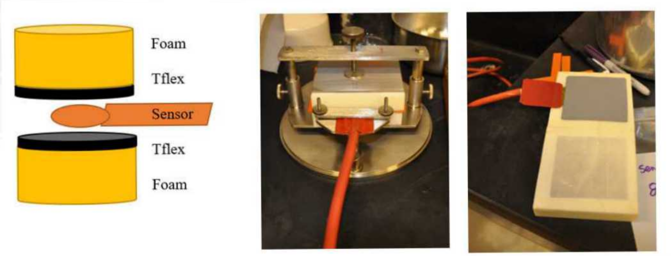
$$\rho c_p = \frac{k_r}{\alpha_r} = \frac{k_a}{\alpha_a}$$

Volumetric specific heat is needed

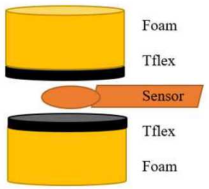
Layered Systems using Thermal Interface Materials

Karla Reyes (PI), Jacob Mayer and Austin Acosta





Layered Systems



Polyurethane foams

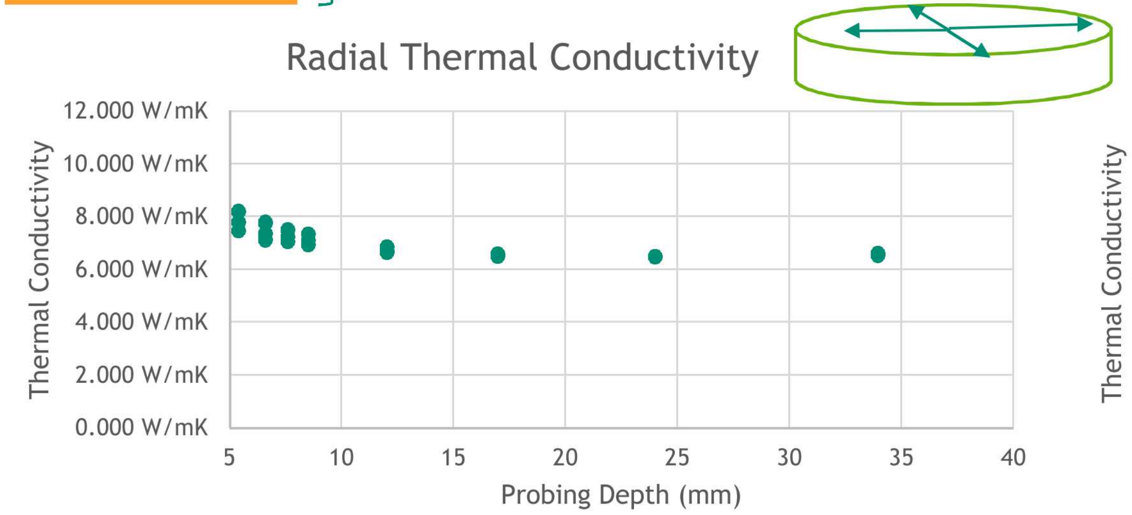
Sample Name	Density (kg/m^3)	Therm Cond. (W/mK)
Last-A-Foam-EF4003	48	0.04019
Last-A-Foam-5020-07	112	0.04452
Last-A-Foam-6070-8.5	136	0.04589

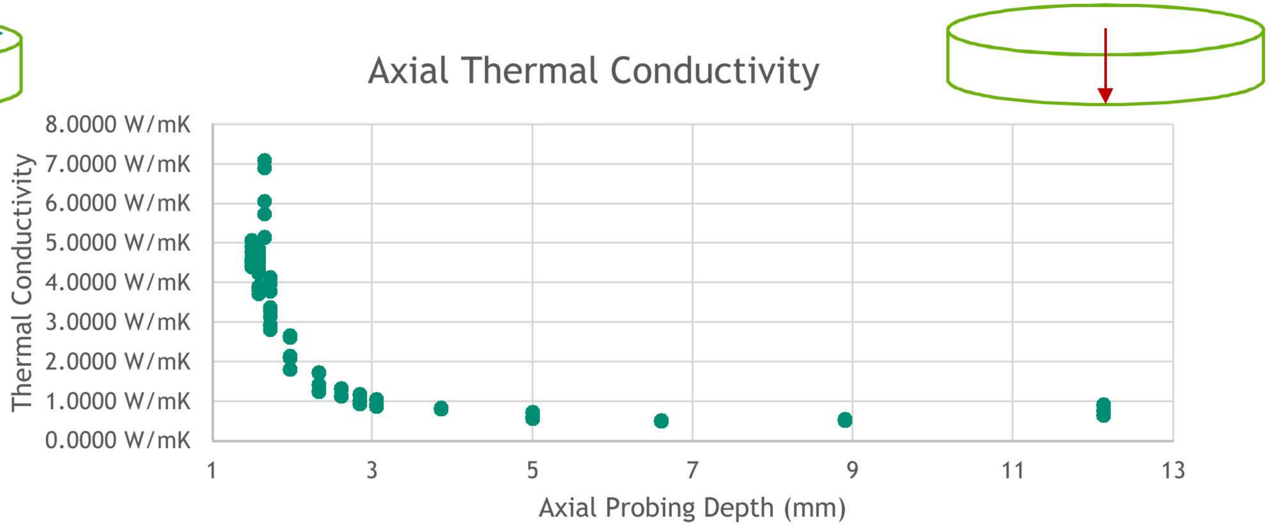
Tflex – 1.00 mm

Polyurethane foams – 23.63 mm

Tflex

- Tflex™ 700 is a soft gap filler thermal interface material with great thermal performance and high compliancy.
- Unique silicone and ceramic filler technology
- Tflex™ 700 is electrically insulating, it is stable from -45°C thru 200°C and meets UL 94V0 flame rating.
- Naturally tacky, it requires no additional adhesive coating to inhibit thermal performance.
- Thermal conductivity of Tflex is 7.1 mW/K





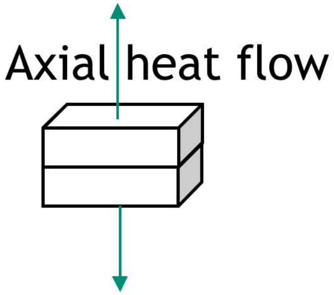
The thermal conductivity was measured at different measuring time (0.1, 0.2, 0.3, 0.5, 1, 3, 5, 10, 20, 40, 80 and 160 s). Probing depth was determined using the thermal diffusivity and specific heat using the rule of mixture.

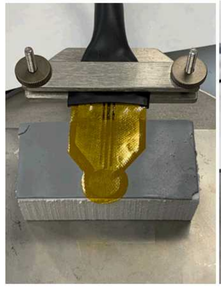
Layered Systems→ Thermal Images

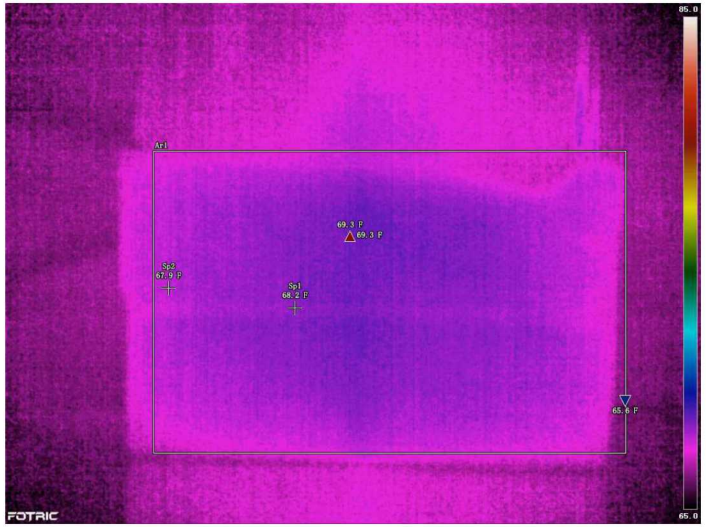
Hot disk sensor in between two pieces of polyurethane foam. 100 mW applied for 80 s

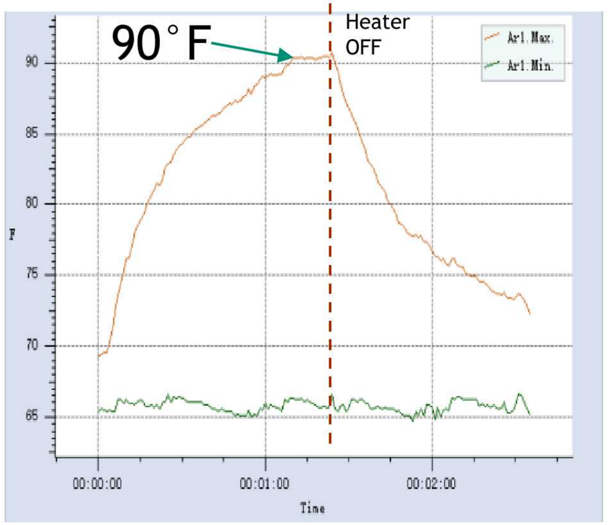
Experiment #1: Bare foam
- ΔT > 20 degrees
- concentrated in the center
- isotropic heating dissipation
- partial heat dissipation after 60 seconds.

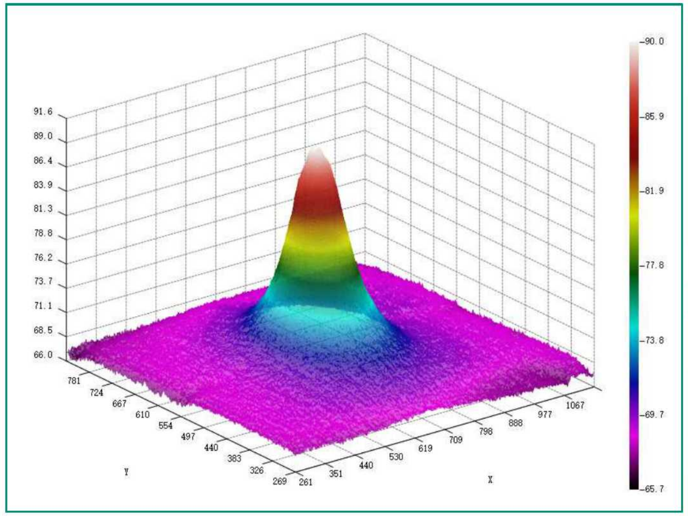
Experiment # 2: Tflex/foam
- ΔT < 5 degrees
- Radial heating dissipation
- Full heat dissipation after 60 seconds.

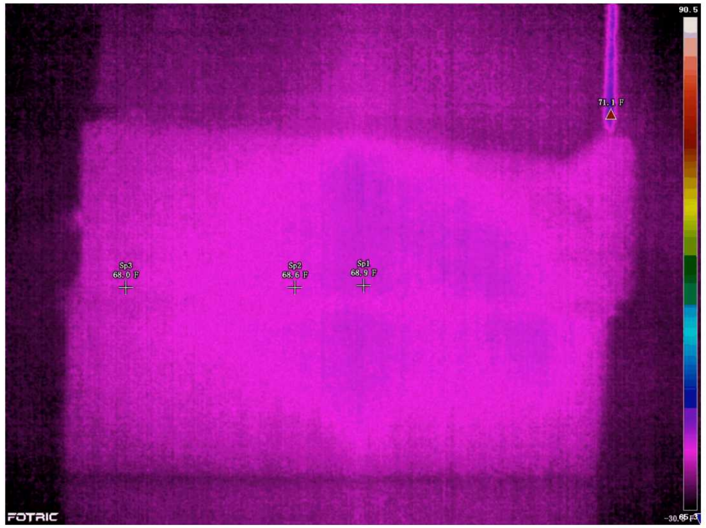


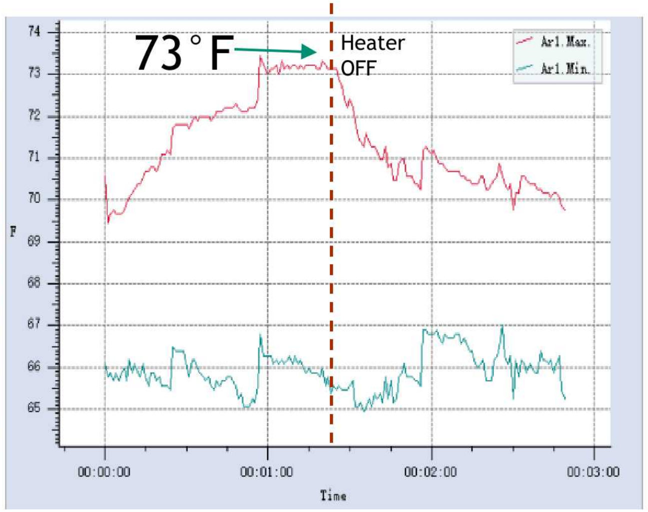


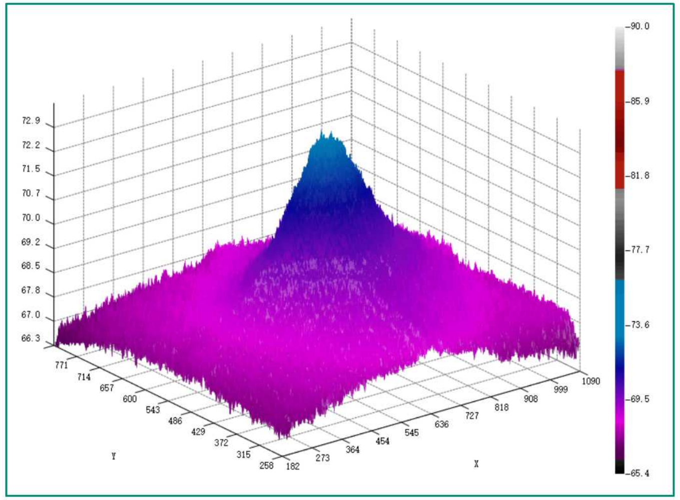












Click on videos

Experimental measurements of 1, 2 and 3 layers

1 layer

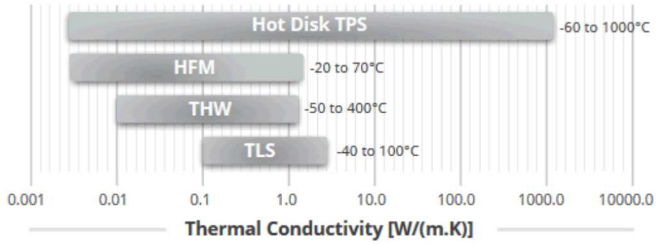
Aluminum (~160W/mK)

Tflex (~5 W/mK)

Foam (0.02-0.04 W/mK)

Sample Name	Therm Cond. (W/mK)	Std. Dev
Last-A-Foam-6070-8.5	0.046	0.004
Last-A-Foam-EF4003	0.040	0.002
Last-A-Foam-5020-07	0.045	0.002
Polyethylene	0.022	0.002

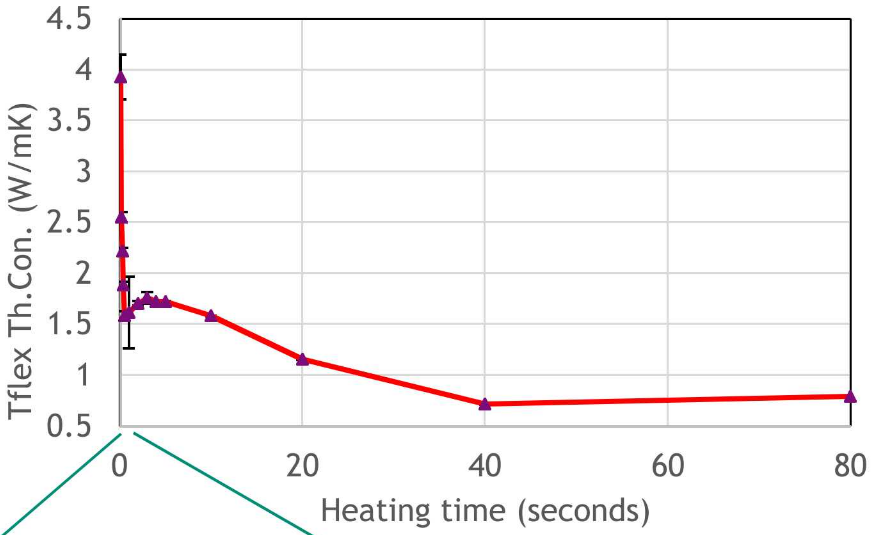
These results show the wide range of Thermal Conductivities the Hot Disk equipment can measure from highly conductive to extremely insulating!

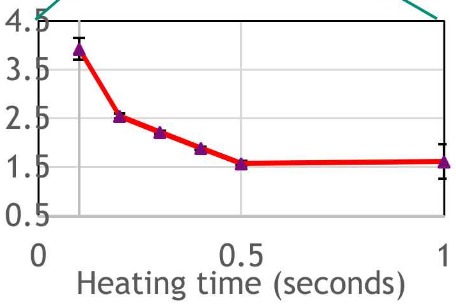


2 layers

Tflex (5 W/mK)

Foam (0.02 W/mK)





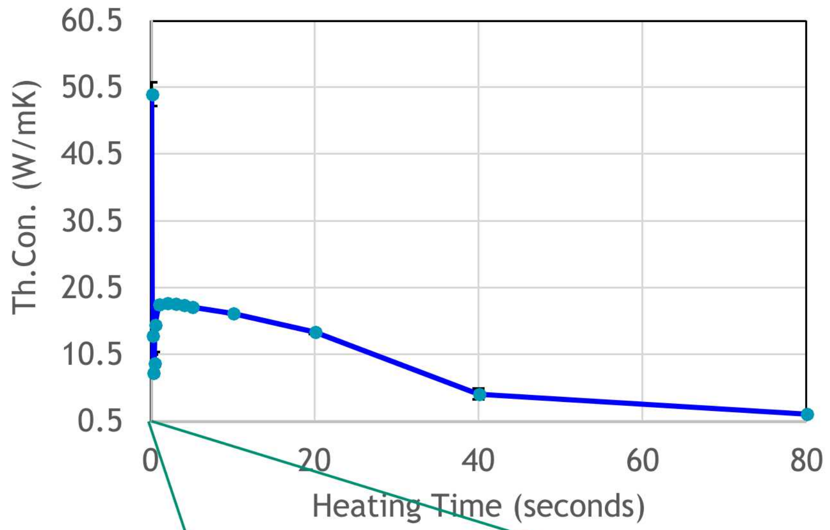
The heat wave quickly travels through the first high conductive layer, as we see the thermal conductivity rapidly decreasing. The Hot Disk equipment has the time resolution to detect these fast transitions!

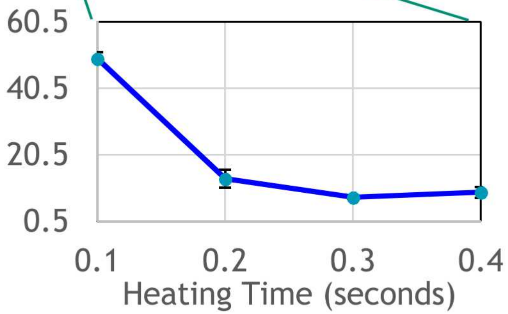
3 layers

Aluminum (0.61 mm)

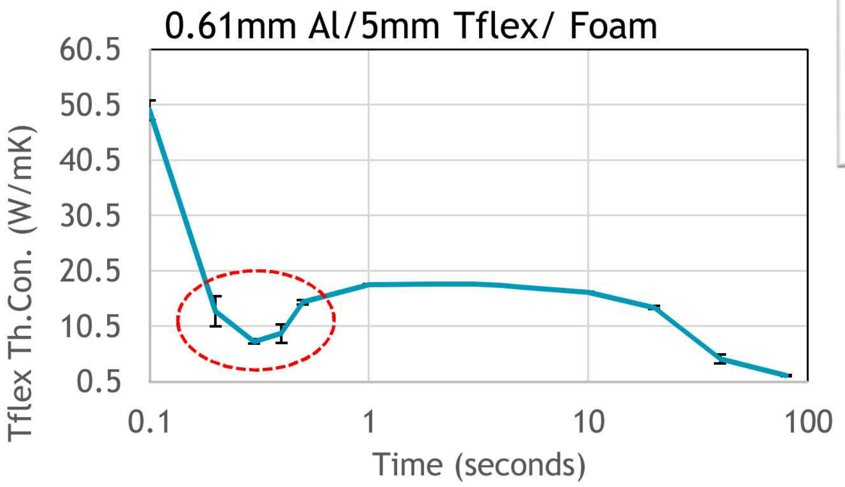
Tflex (5 mm)

Foam (23 mm)





Thermal Contact Resistance



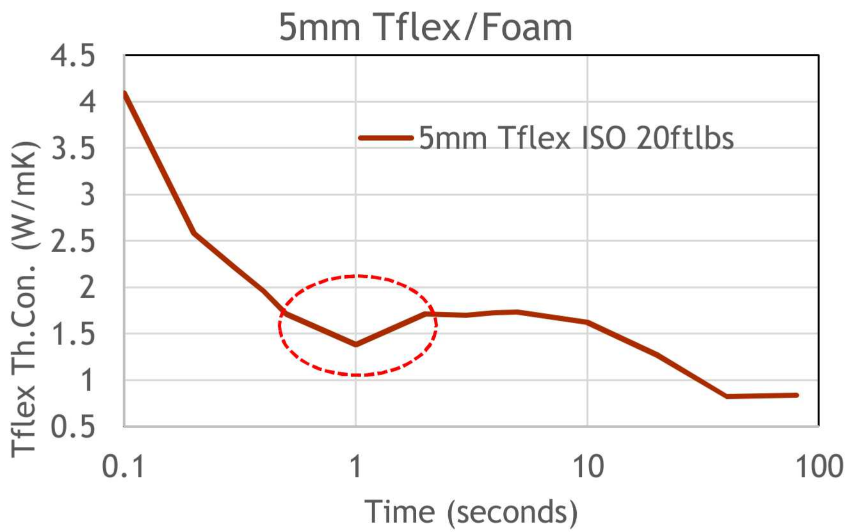
$$R_{\lambda_m} = R_{\lambda_b} + R_c \longrightarrow \frac{d}{\lambda_m} = \frac{d_b}{\lambda_b} + R_c$$

Where d = distance = thickness
m = measured
b = bulk
c = contact

On the left plots (under 20 ft lbs), the contact resistance can be seen as reduction on thermal conductivity when the heat wave reaches the interface between the two layers.

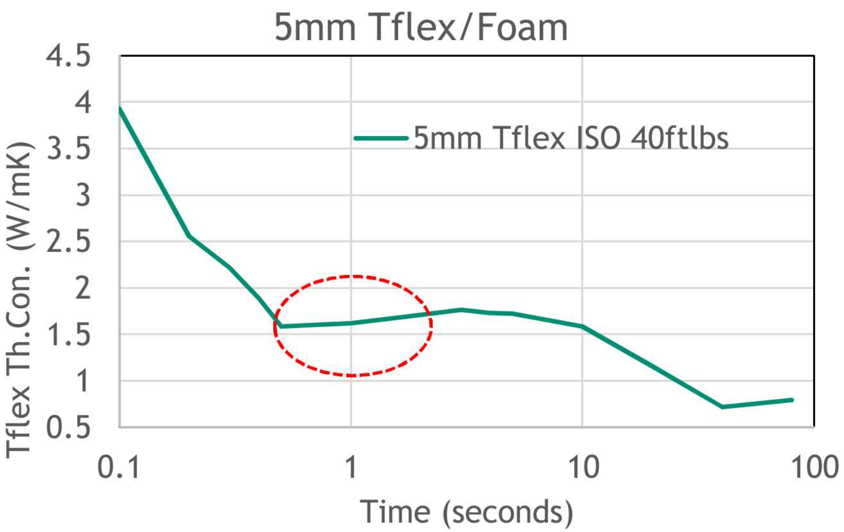
On the right plot (under 40 ft lbs), the contact resistance is reduced. The higher applied load improve the contact between the two layers, reducing air gaps.

20 ft lbs applied



Compression stand use to applied different load to the samples.

40 ft lbs applied



Thickness Dependency on Thermal Conductivity

At short times, the first layer is going to dominate the overall thermal conductivity.

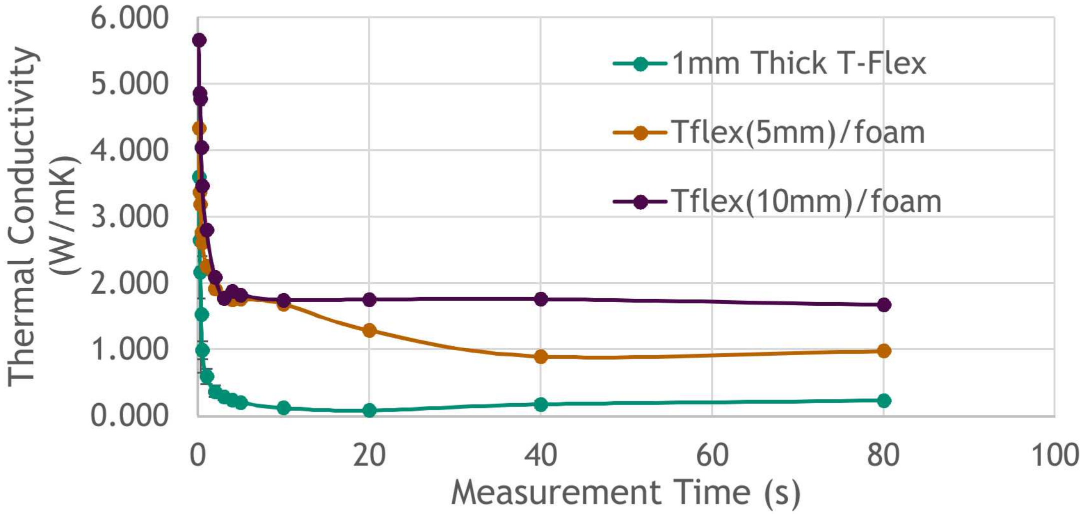
At longer times, the thermal conductivity is going to be a weighted average of the thermal conductivities of the different layers.

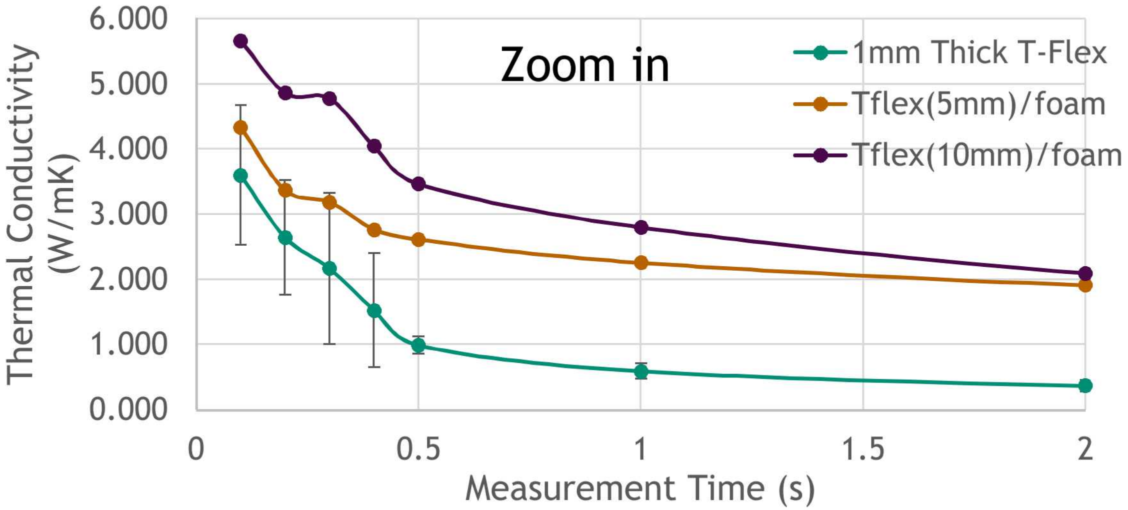
The thickness dependency results show that by changing the thickness of the highly conductive material the overall thermal conductivity can be changed.

As shown in the “zoom in” plot, the rate of thermal conductivity decrease directly depends on the thickness of the Tflex (the high conductive layer)

Thickness dependency on the thermal conductivity

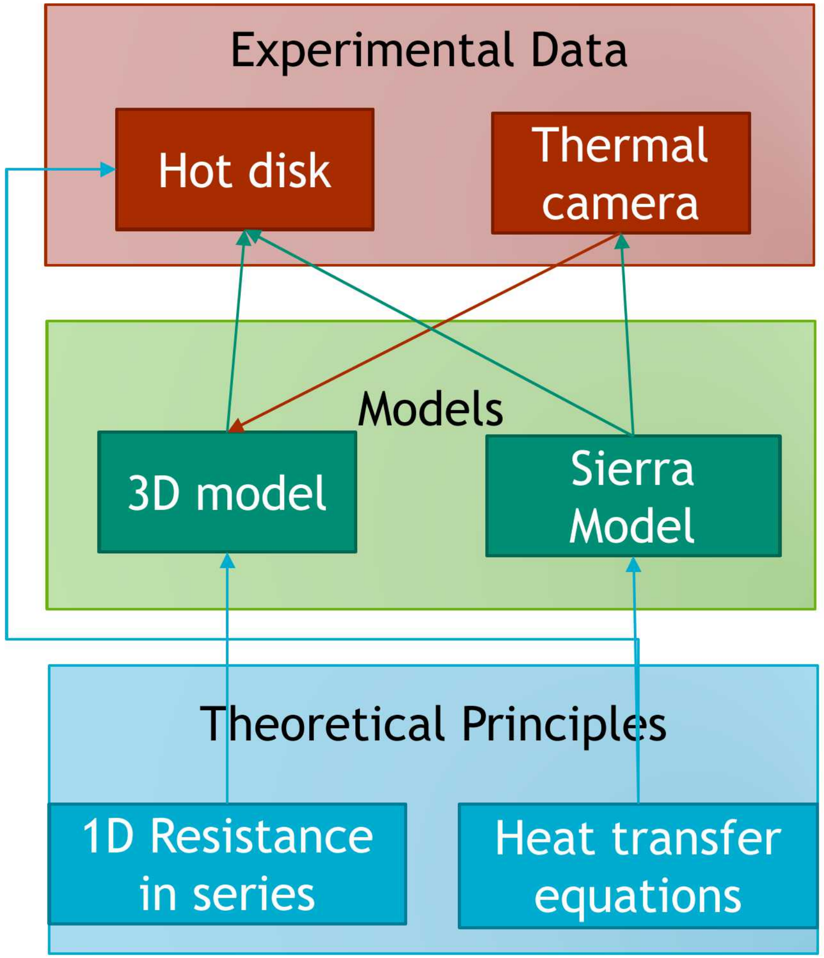
- 1mm, 5mm and 10mm T-Flex/Foam





Validation Process Flow and Prediction Tool Development

To validate experimental data, we used theoretical principles to compare the experimental data. Then we used the principles to build models. Finally we fed the models with experimental data to make them more realistic.



Experimental Data

- Hot Disk → Thermal Conductivity values as function of measuring time
- Thermal Camera→ Temperature Profile as function of time

Models

- Sierra models→ visualization of heat flow
- Modified 3D model→ use the resistance in series principle applied to a 3D systems

Theoretical Principles

- Resistance in series → thermal conductivity in 1D direction
- Heat flow equations→ transient plane source

Thermal images vs simulated heat flow

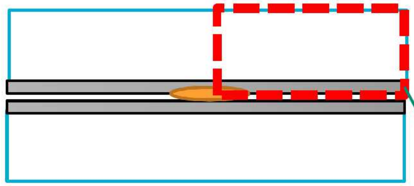
Thermal camera ⟷ **Sierra Model**

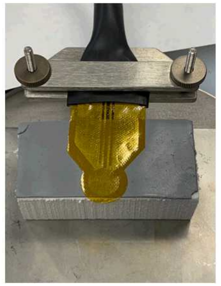
Heat flow (experimental vs. simulated)

Independent methods

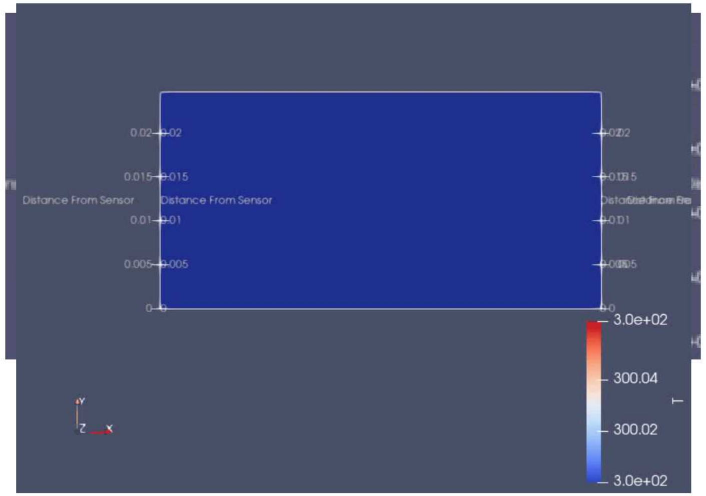
Experimental parameters affecting thermal conductivity are contact resistance, pressure, surface treatments, instrument and human errors (not included in the simulations)

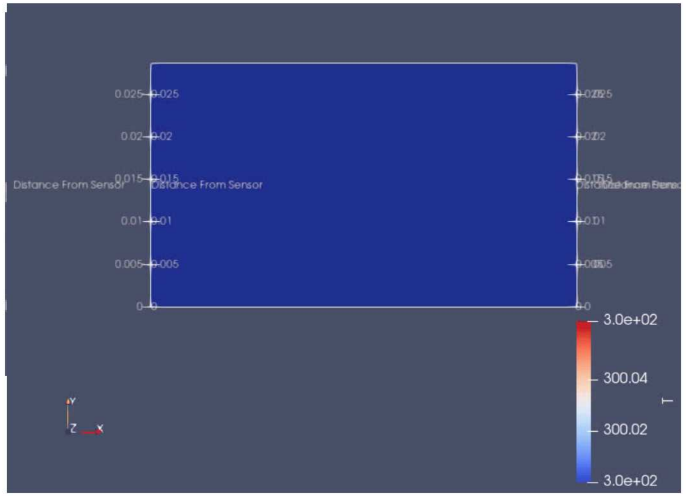
Based on heat flow theory

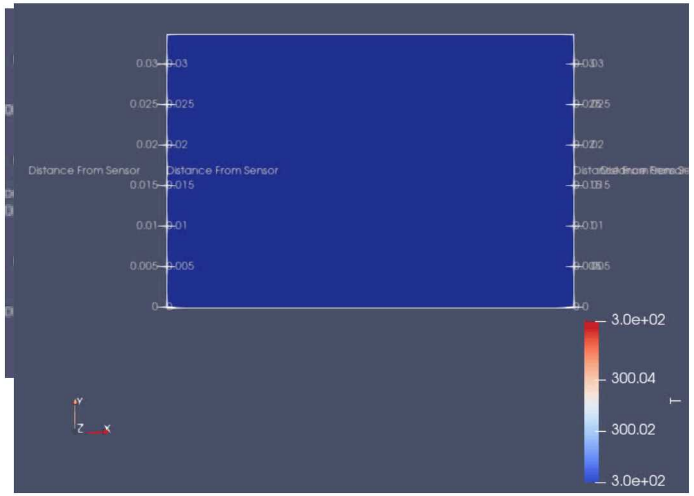


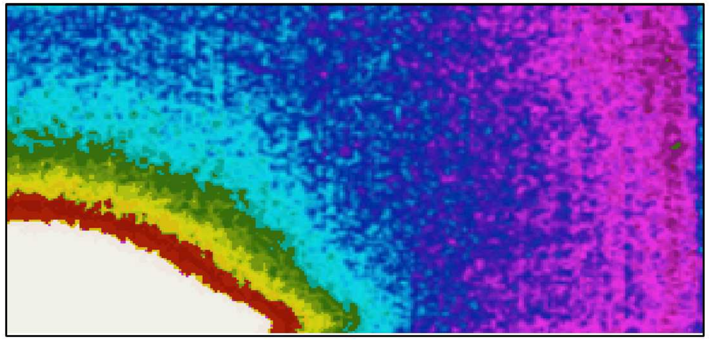


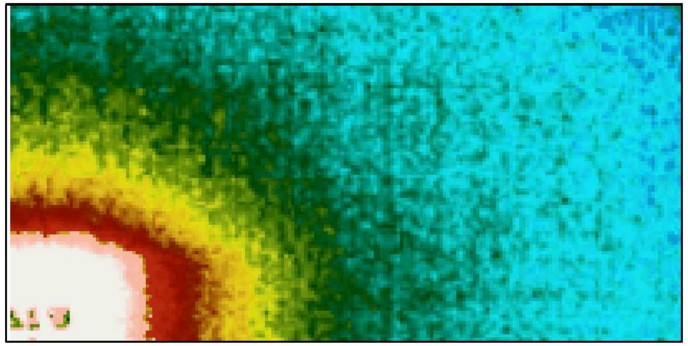
Right quadrant image, the heat source is in the left bottom corner

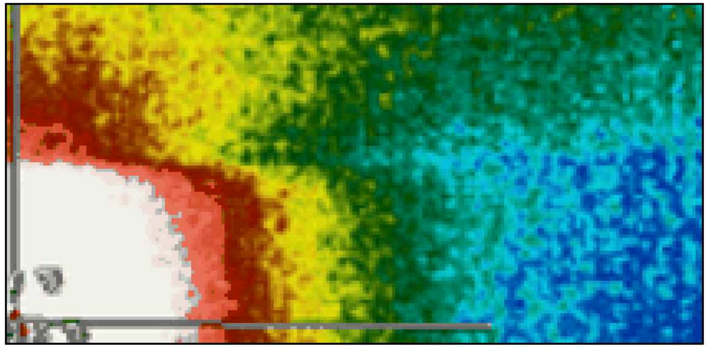












1D Resistance in Series

Hot disk ⟷ **1D Resistance in series**

Thermal Conductivity (experimental vs. simulated)

Independent methods

Experimental parameters affecting thermal conductivity are contact resistance, pressure, surface treatments, instrument and human errors

Theoretical Principle Assuming 1D heat transfer

- Thermal resistance is a measurement of a temperature difference of a material's ability to resist the flow of heat. It is often described as the reciprocal of thermal conductance.
- This thermal property greatly depends on the area, thickness, and thermal conductivity of a material. Resistance increases by increasing the thickness of the material, and decreasing its area and thermal conductivity.
- Thermal resistance can be compared to an electrical circuit model. In this case, heat flow is represented by current, temperatures are substituted with voltages, and resistances are expressed as resistors.
- The thermal resistance of a whole object (like a wall) can be calculated from the resistance of each individual layer. All layers should have the same area, but they can have different thicknesses. When the heat flux through multiple mediums is assumed constant, the thermal resistance "in series" can be determined.
- This does not account for contact resistance between the different layers or components. Effects like this should be directly measured.

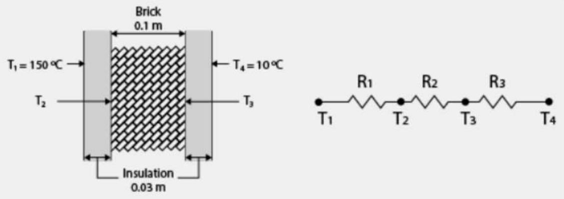


Figure 3. The thermal resistances (R) and temperatures (T) of a wall surrounded by insulation (left), modelled as an electrical current (right).

Thermal Resistance:

$$R = x/(A * k)$$

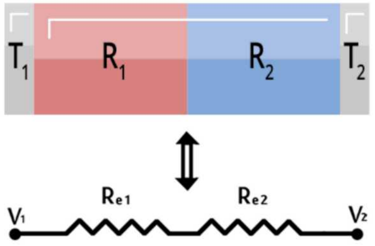
Where:

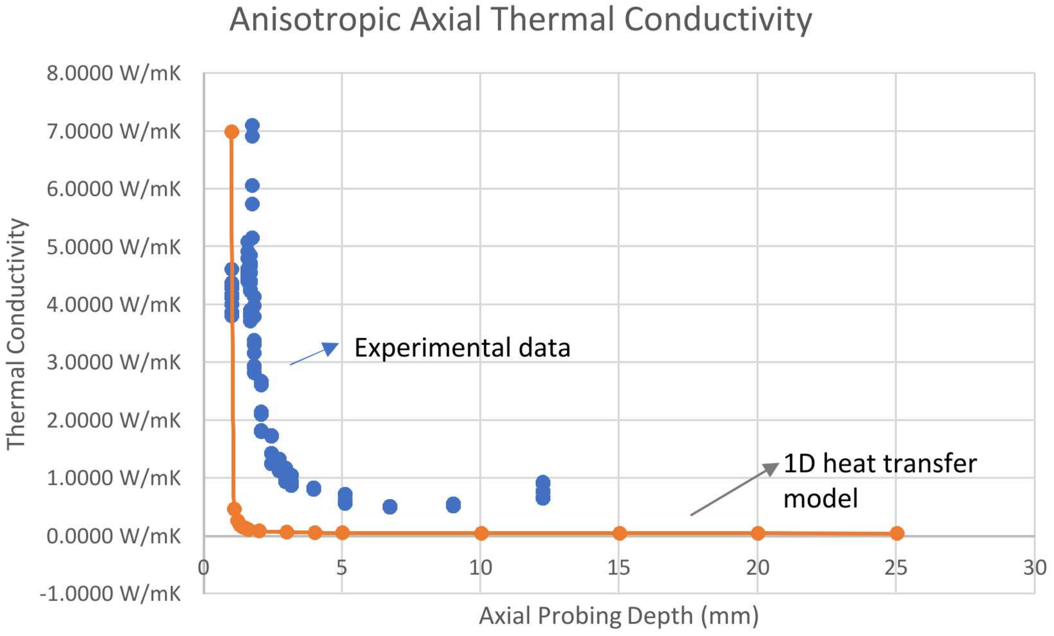
R = thermal resistance

x = penetration depth or width of material

A = area

k = thermal conductivity

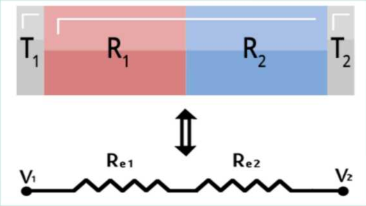




http://web.mit.edu/16.unified/www/FALL/thermodynamics/notes/node118.html

ID vs 3D Heat Transfer

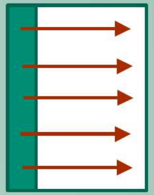
1D heat transfer



$$R = R_1 + R_2 \dots$$

$$R = \frac{x1}{A * k1} + \frac{x2}{A * k2} \dots$$

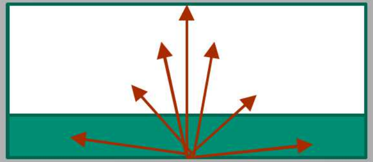
All layers should have the same area (A), but they can have different thicknesses (x). The heat flux through multiple mediums is assumed constant.

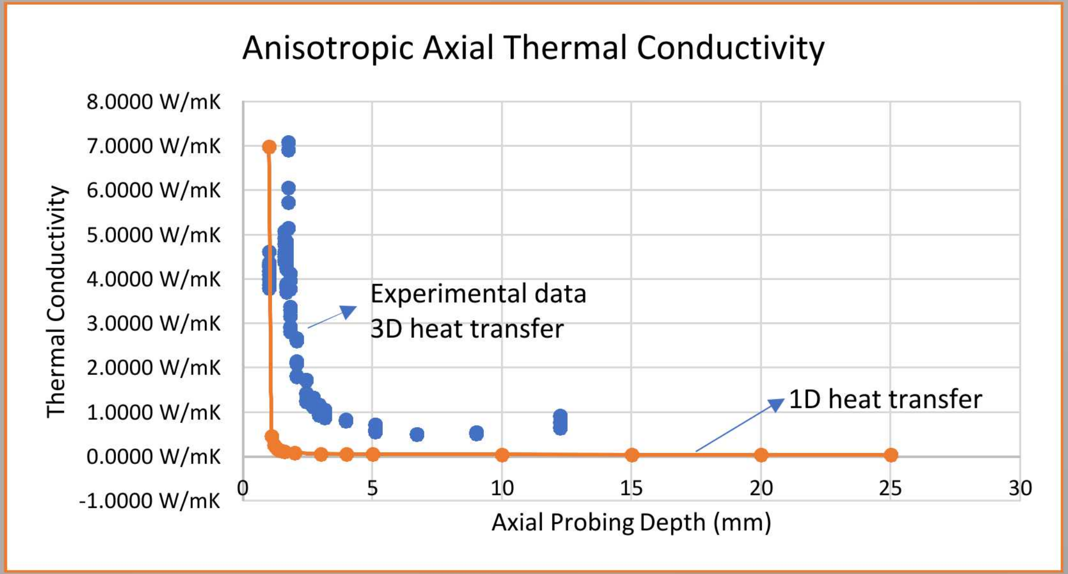


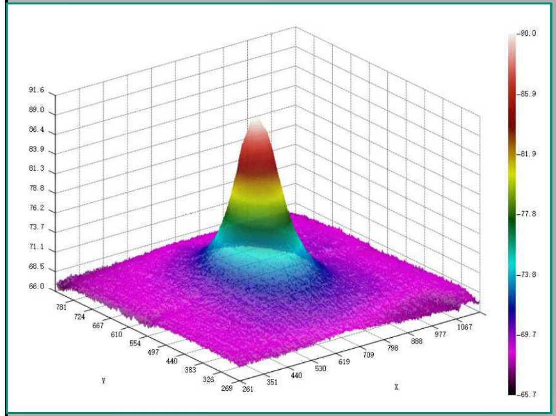
1D assumption: cross sectional area of heat wave is constant through the layers

3D heat transfer model is needed

Problem: Heat wave area through the layers is not constant. More like a gaussian shape





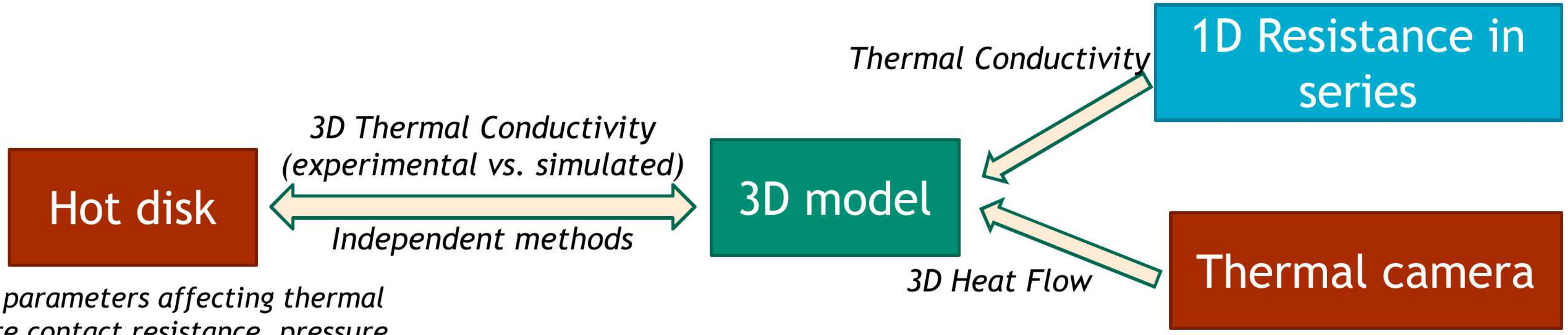


$$R = R_1 + R_2 \dots$$

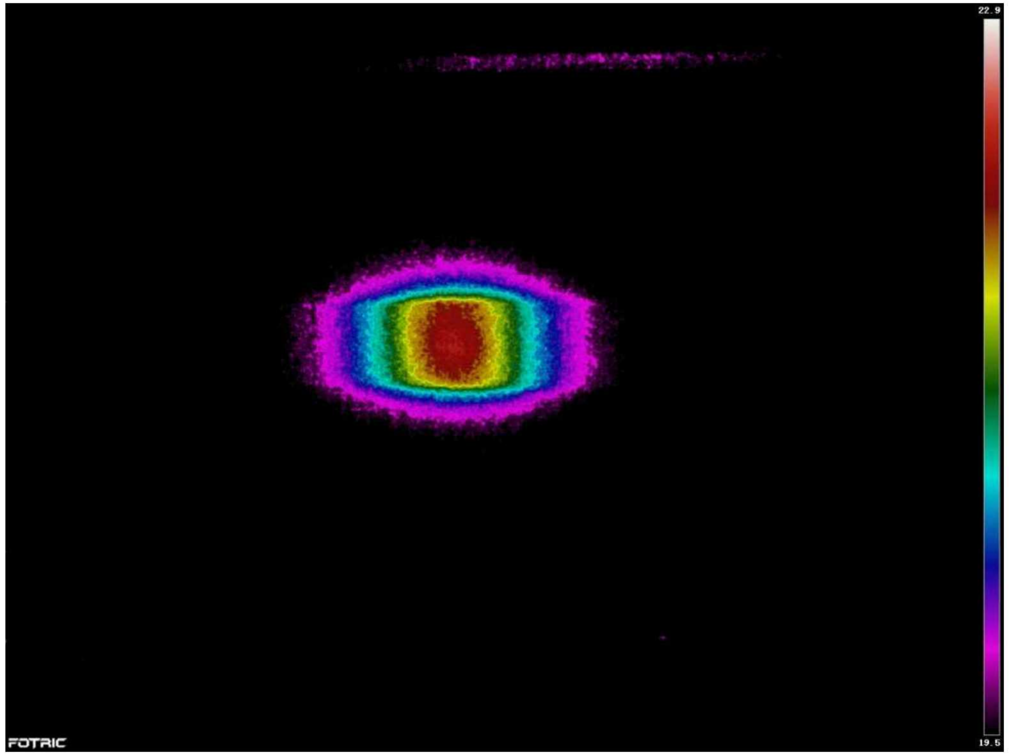
$$R = \frac{x1}{A1 * k1} + \frac{x2}{A2 * k2} \dots \qquad A1 \neq A2$$

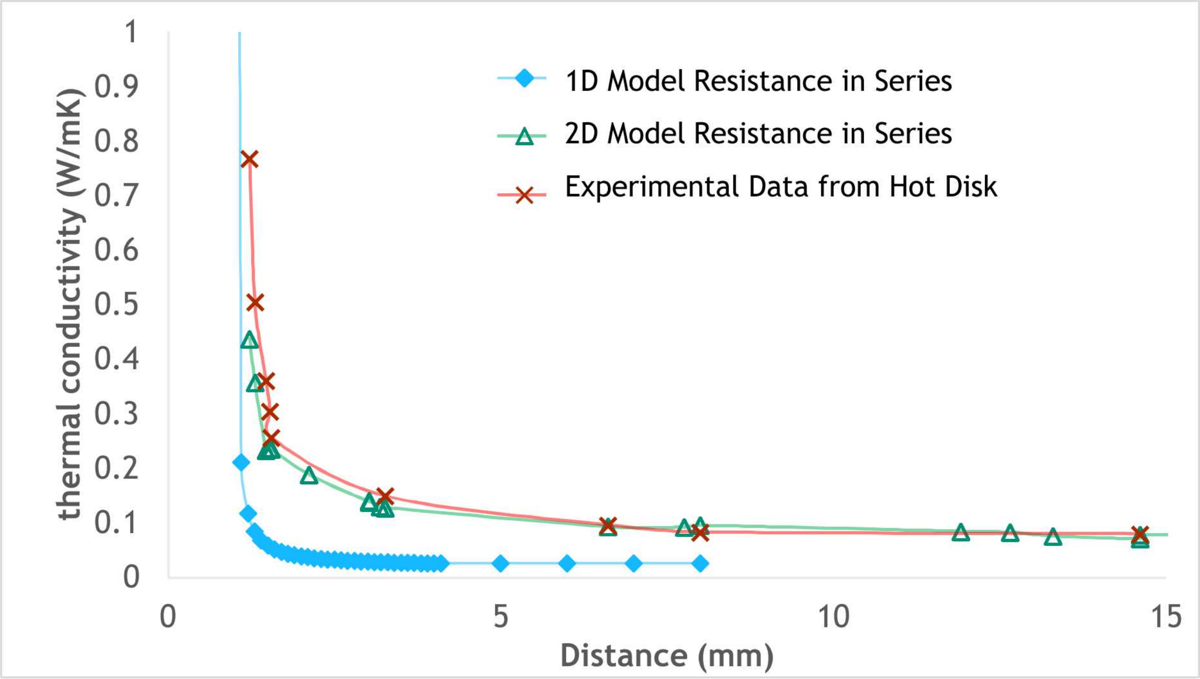
ID model is overestimating A2. The transition from layer 1 (Tflex) to layer 2 (foam) is not a abrupt step, instead of ramp process.

Accomplishment # 10: 3D Heat Flow Model



Experimental parameters affecting thermal conductivity are contact resistance, pressure, surface treatments, instrument and human errors

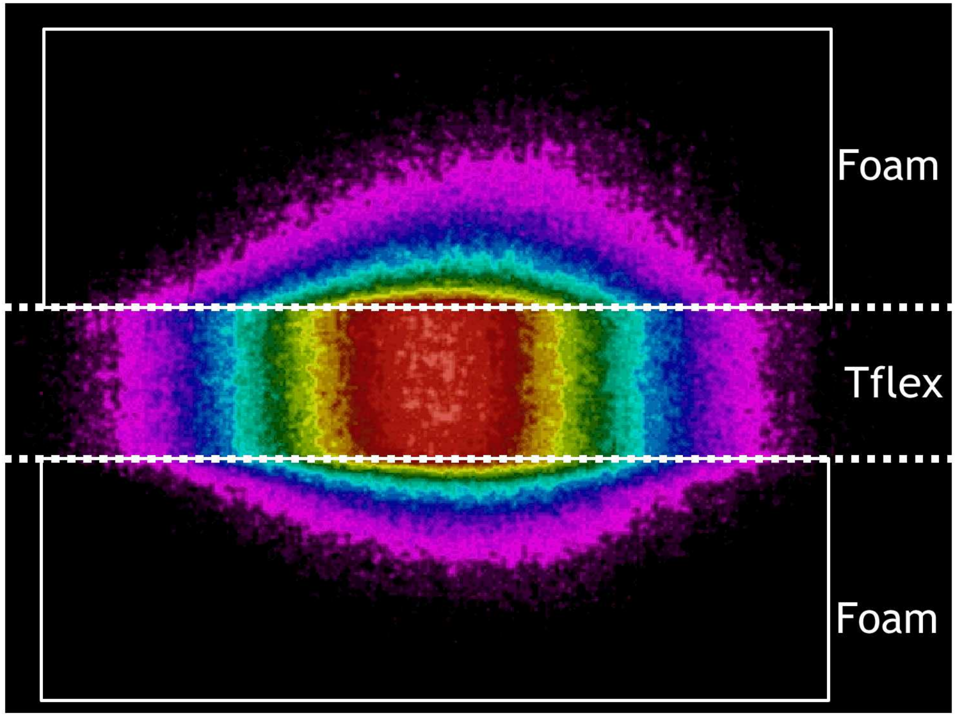




The predictions using the 2D model are in a very good agreement with the experimental data

Watch animation in slide show mode

2D Heat Flow Model in Matlab



$$\int \Delta T_{Foam}\, dA$$

$$\int \Delta T_{Tflex}\, dA$$

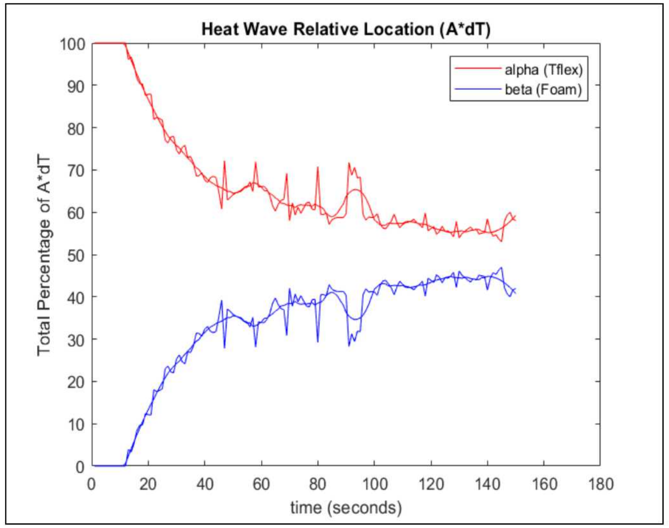
$$\int \Delta T_{Foam}\, dA$$

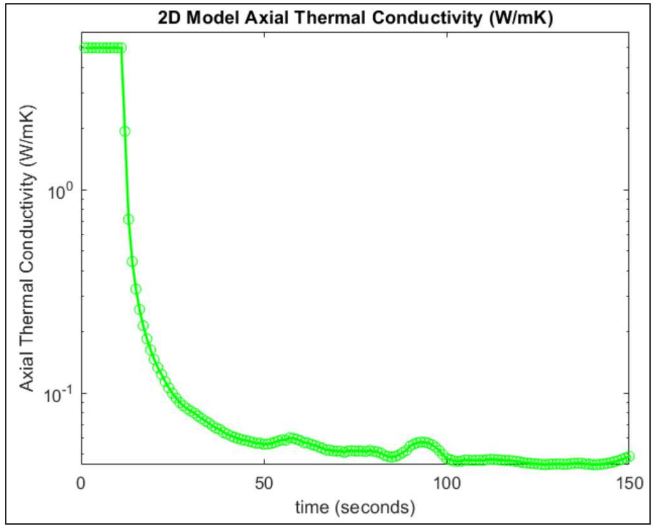
$$k = \left(\frac{\alpha}{k_{Tflex}} + \frac{\beta}{k_{Foam}} \right)^{-1}$$

$$\alpha = \frac{\int \Delta T_{Tflex}\, dA}{\int \Delta T_{Tflex}\, dA + \int \Delta T_{Foam}\, dA} \qquad \beta = \frac{\int \Delta T_{Foam}\, dA}{\int \Delta T_{Tflex}\, dA + \int \Delta T_{Foam}\, dA}$$

$$k_{Tflex} = 5 \left(\frac{W}{m * K}\right) \qquad k_{Foam} = 0.02 \left(\frac{W}{m * K}\right)$$

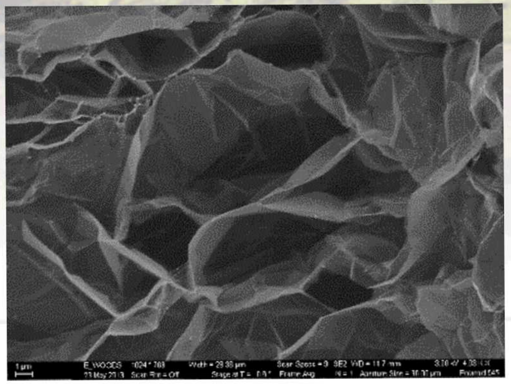
A code in Matlab was created to analyze the heat wave area using the thermal images.
As the heat flow in the x-axis should be very similar to the flow in the y-axis, we want to assume that the 2D heat flow is very similar to the 3D heat flow.

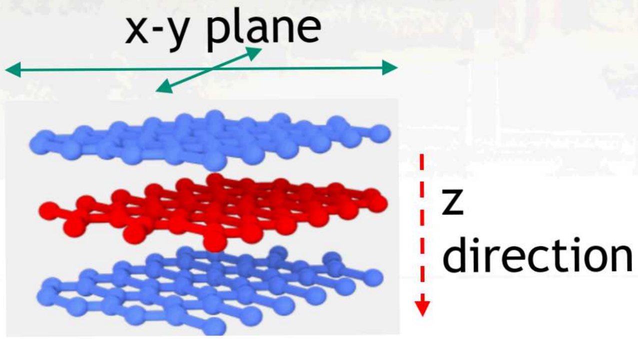


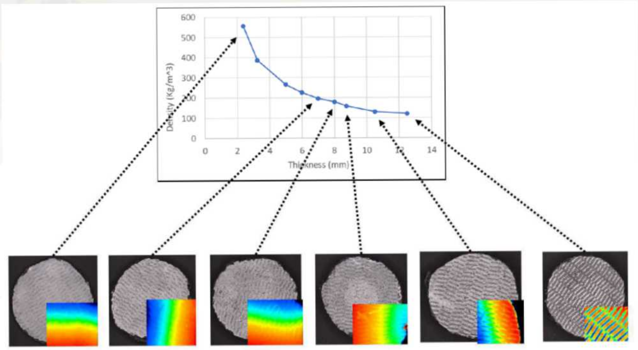


Compressed Graphite

Karla Reyes (PI), Jacob Maher, and Anne Mallow

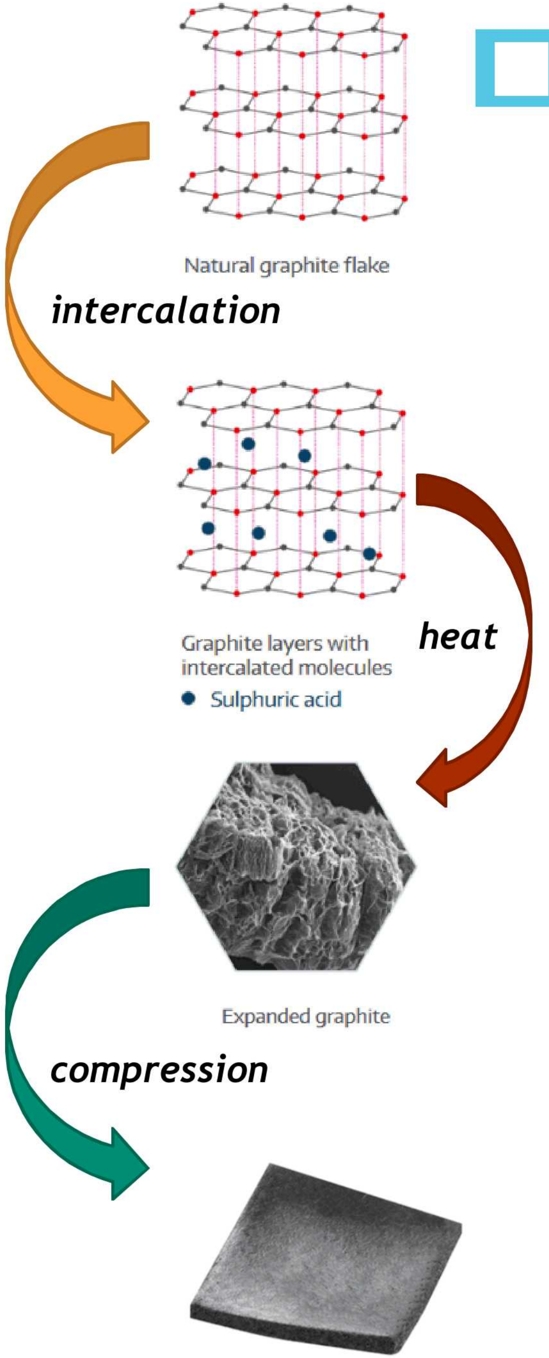




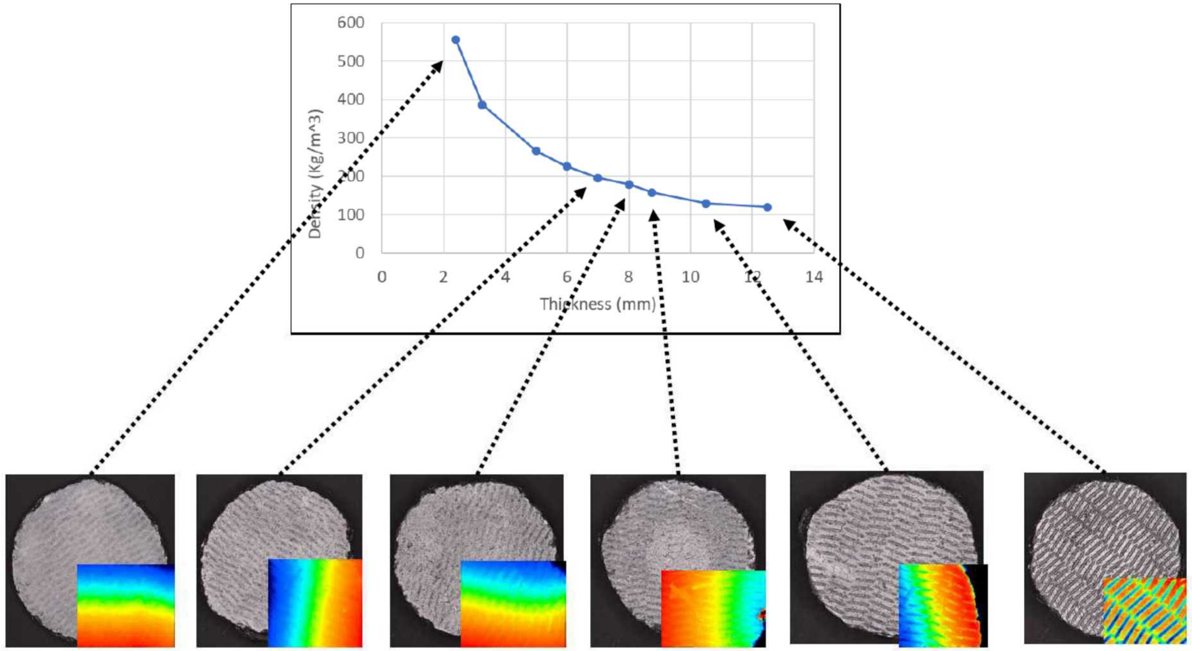


Compacted Expanded Graphite

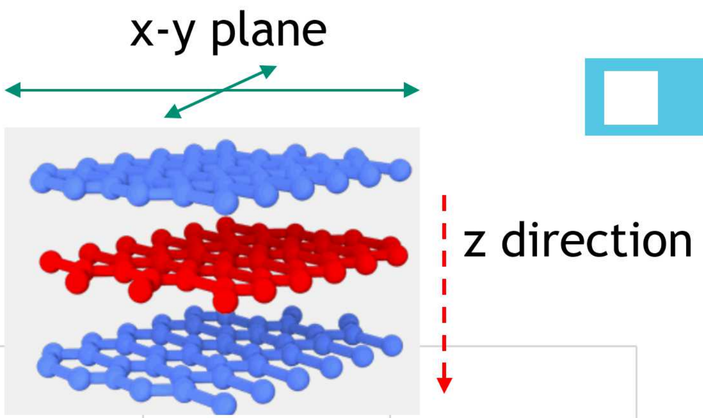
- Due to the layered structure of graphite, atoms or small moelecules can be intercalated between the carbon layers producing graphite intercalation compound (GIC). Usually Sulphur or Nitrogen are used as intercalation agents
- Under the effect of heat the layers separate like accordion and the graphite flakes expand.
- The final volume can be several hundred times greater than the initial volume.
- Expansion of the flake graphite decreases the material's overall bulk density while increasing its surface area. Expandable graphite is valued as a flame retardant. Expansion creates an intumescent layer on the surface of the graphite flake that slows the spread of fire and minimizes the creation of toxic fumes and gases.
- The graphite is then compressed using different loads to create materials with different densities.

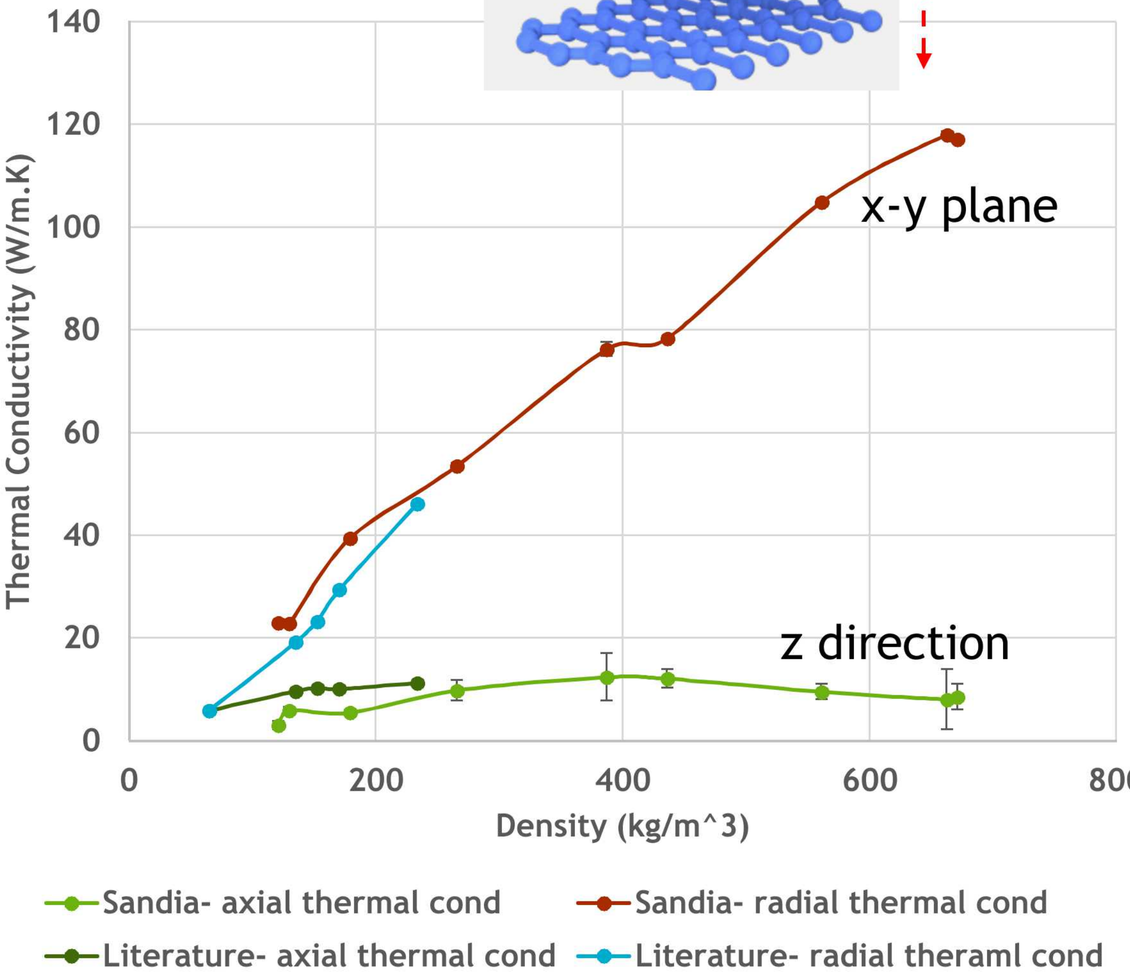


Anisotropic Thermal Conductivity of Graphite



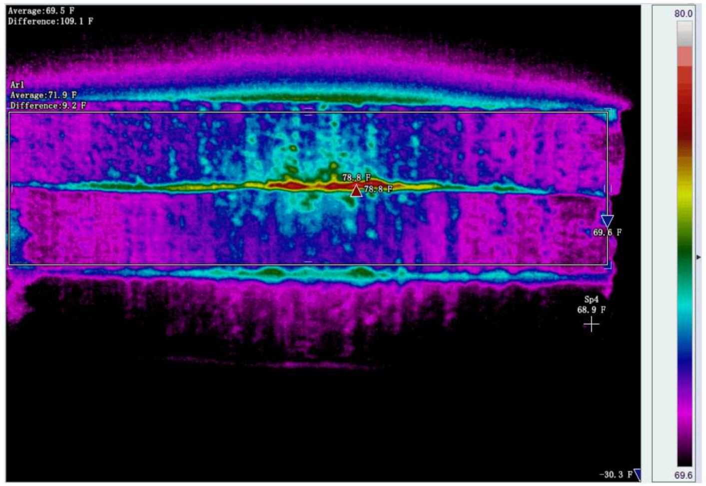
- The graphite was compressed using different loads to create a wide range of densities (100 to 600 kg/m³).
- Good agreement with the literature (Bodzenta, et. al, "Thermal properties of compressed expanded graphite: photothermal measurements", Appl Phy B, 2011, 105, 623-620)
- The radial (x-y plane) thermal conductivity increases proportional to the density.
- The axial (z-direction) thermal conductivity didn't significantly with density.



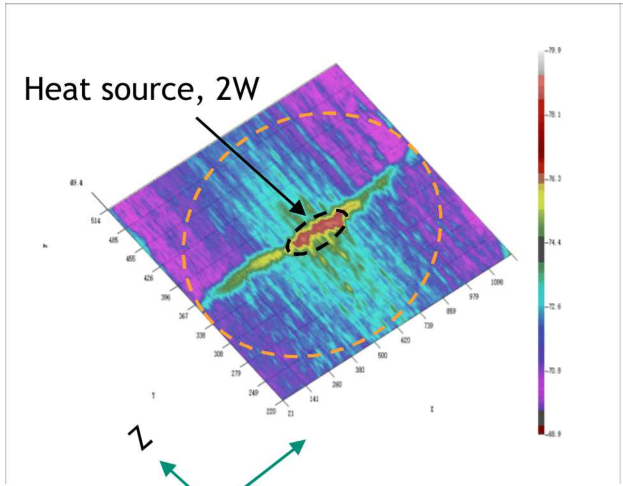


Thermal Images- Uncompressed vs Compressed Graphite

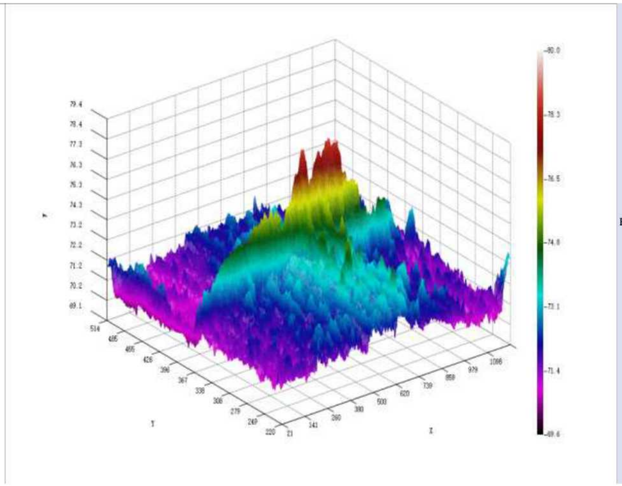
Uncompressed graphite → less dense



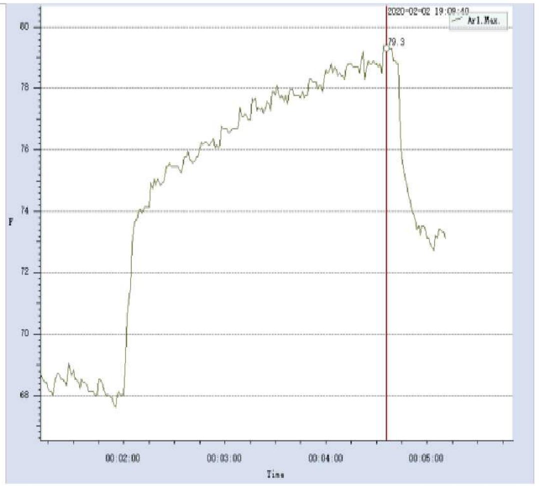
isotropic heat distribution

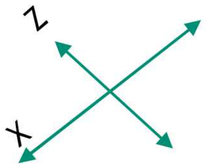


heat concentrates in the center

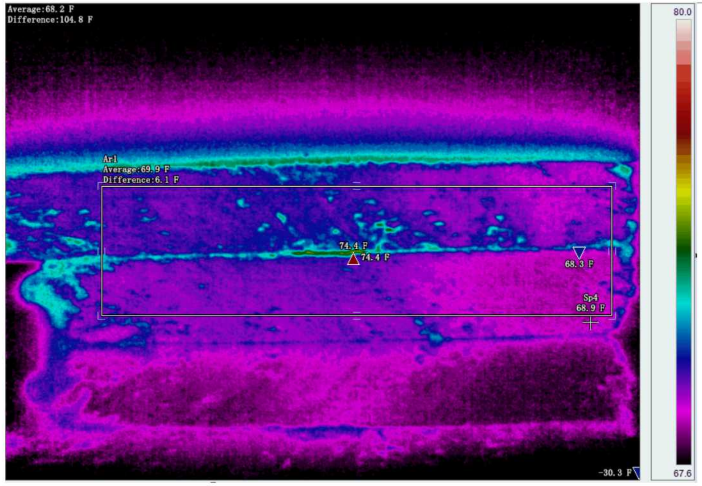


Temp max= 79.3 °C

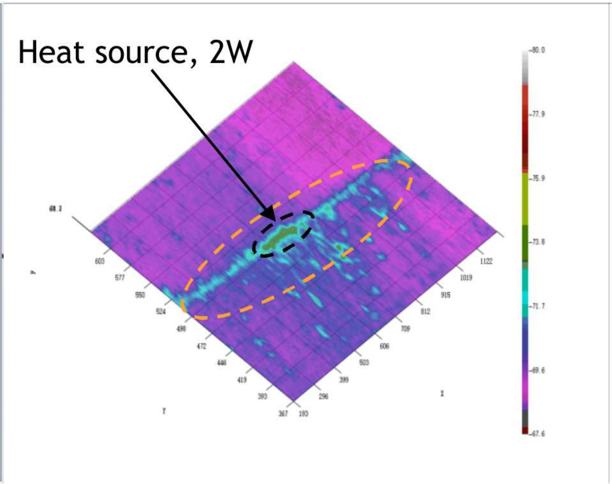




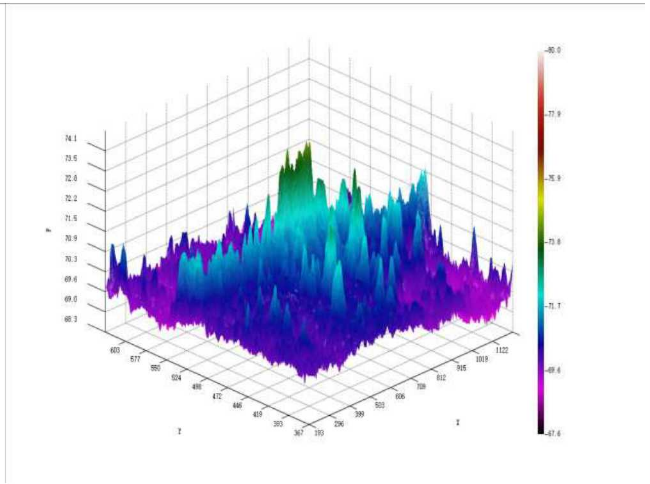
Compressed graphite → more dense



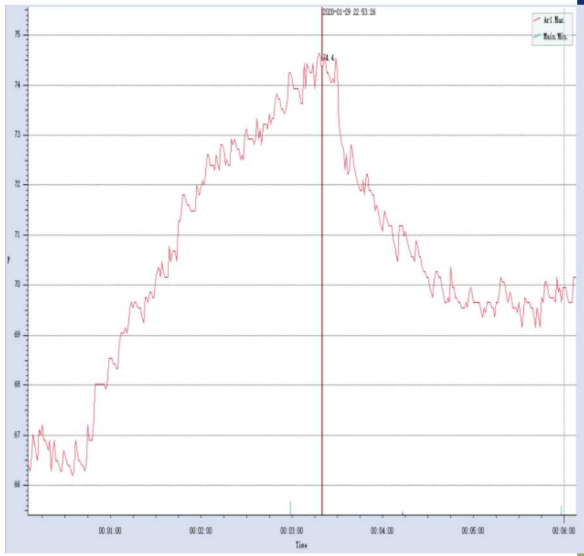
anisotropic heat distribution



heat dissipates from the center

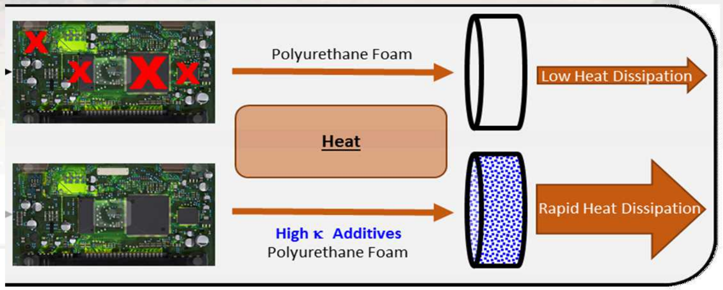


Temp max= 74.4 °C

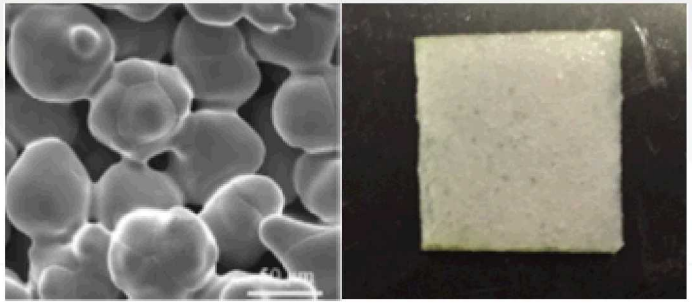


Composite Polyurethane Foam Materials

Matthew Walter (PI), Karla Reyes and April Nissen

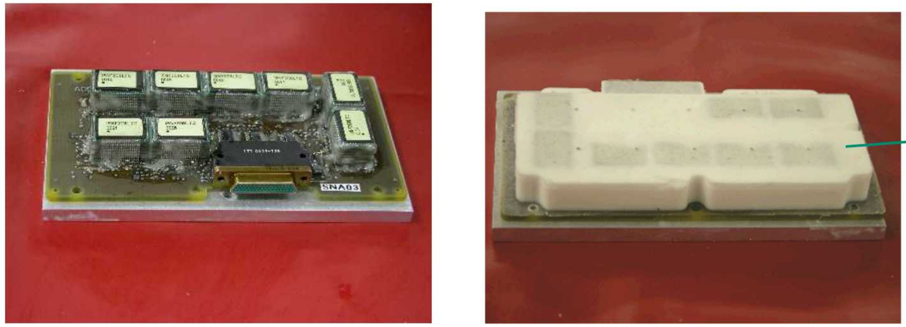


Composite Foam Fabrication

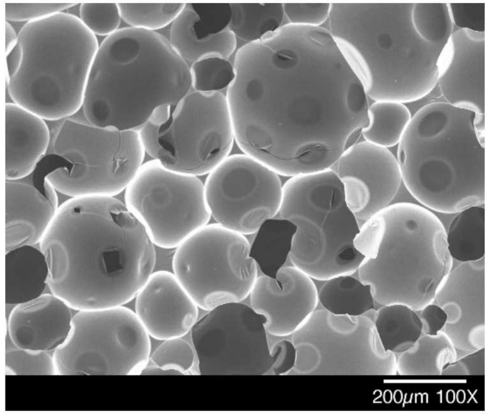


Composite Polyurethane Foam Materials

- Rigid, closed-cell polyurethane foams are used to encapsulate electronics
  - Protect from vibration, shock, and impact

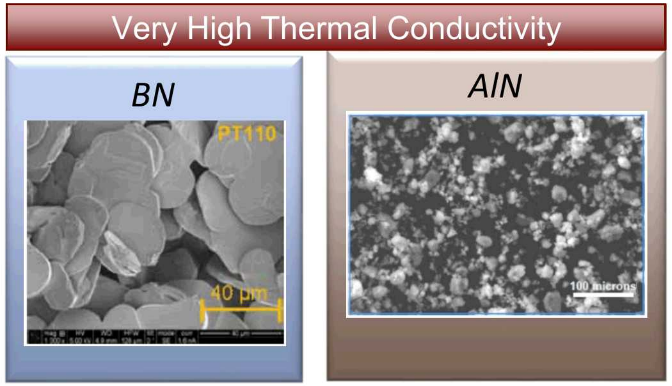


Electronics Foam Encapsulation



Closed-Cell Foam Microstructure

		Aluminum oxide (Al_2O_3)	Boron carbide (B_4C)	Amorphous boron (B)	o-Carborane, 98%	Boron nitride (h-BN)	Aluminum nitride (AlN)
Thermal Properties	Thermal Conductivity (W/m-K)	30	28	27	-	***>300 (IP); 3 (TP)***	***260***
	Specific Heat (J/kg-K @25°C)	798	945	1294	-	794	734
Electrical Properties	Dielectric Constant	9.7	8	-	-	3.9	8.8
	Volume Resistivity (ohm-cm)	10^{14}	10^{3}	10^{6}	-	10^{15}	10^{14}
Physical Properties	CTE (ppm/K)	6.7	5.54	-	-	<1	4.4
	Young's Modulus (GPa)	340	450	-	-	40	400
	Density (g/cm³)	3.98	***2.51***	***2.35***	***0.95***	***2.25***	3.26
Others	Cost ($/kg)	-	***44***	685	***70,000***	180	140
	^{10}B Content (atoms) - 1 wt% in 14pcf Foam	0	***3.06E+22***	***3.92E+22***	***3.16E+22***	***1.70E+22***	0



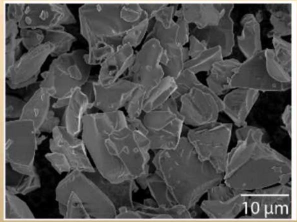
Fabricating Composite Foams Over Wide Density Range

Experimental Overview

Composite Foam Matrix

- Density

Additive Materials



- Material
- Particle Size
- Loading (wt %)

Composite Foam Fabrication

B_4C AlN BN Amorphous B

Foams prepared by incorporating dried additives into the resin mixture, adding isocyanate to the mixture, and then casting in a cylindrical aluminum mold

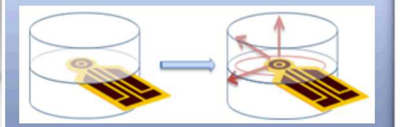
Physical Properties

- Density
- T_g
- Storage Modulus

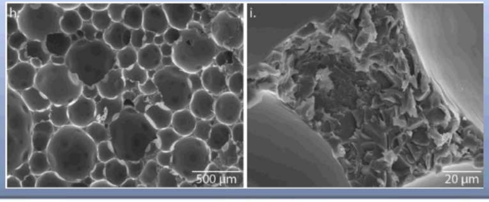
Neutron Shielding Properties

- Neutron Attenuation
- Secondary Ionization Products

Electrical / Thermal Conductivity

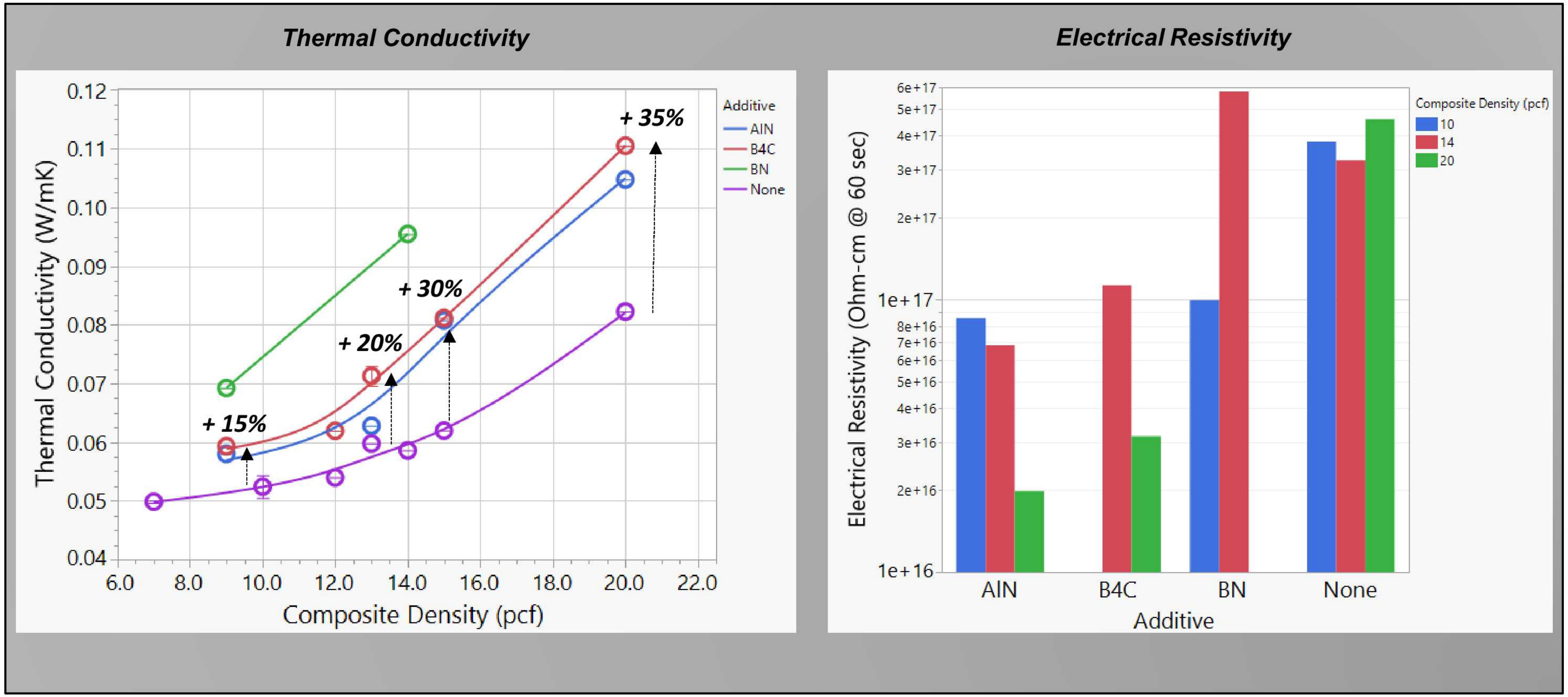


Microstructural Characterization



Thermal Conductivity Significantly Increased by Additives

Thermal Conductivity and Electrical Resistivity



- **Significant increases in thermal conductivity (15-35%) observed for B_4C composite foams**
- **BN composite foams had the highest thermal conductivity, but are extremely difficult to process**
- **Additive loaded composite foams maintain high electrical resistivity (values typical of electrical insulator)**

Future work: Nanostructured Composites

Approach # 1: Incorporated nanostructured additives to foams as filler materials to increase the thermal conductivity of dielectric polymeric composites.

- One-dimensional (1D) structures can create one-directional thermal pathways to dissipate heat, including carbon nanotubes, hexagonal and cubic phases of BN, graphene and graphite foams
- By using nickel template formed by sintered nickel powder rather than the traditional reticulated nickel foam templates, the pore size of the foams decreases and thus the specific surface area increases allowing for better thermal conductance and greater density.

Approach # 2: Electrochemical co-deposited coatings

- This process enables the growth of a composite coating in a single step. This mechanism entails the electrochemical reduction of a matrix phase, typically a metal, while a second particulate phase is incorporated through entrapment in the growing film.
- Low thermal conductivity materials will be incorporated to modify heat transfer properties of the films produced.

Approach # 3: High void fraction AM metallic structures

- Control porosity fraction within a metallic surface layer fabricated by additively manufactured (AM)
- Low thermal conductivity materials will be incorporated in the voids.

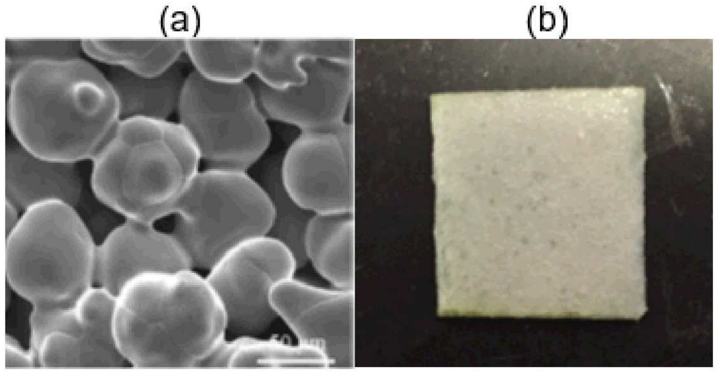


Figure 1(a) Scanning electron micrograph of sintered nickel foam catalyst (b) optical micrograph of an 11 mm x16 mm x 1.6 mm h-BN/PMMA foam composite.

Images courtesy of :
Prof. Li Shi, University of Texas at Austin

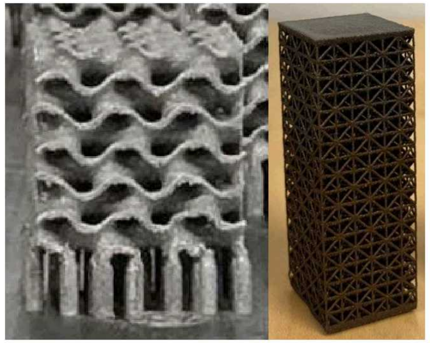


Figure 2: Sample AM gyroid (left) and lattice (right) structures. *Images courtesy of Prof. Mark Hoffman, Auburn University.*

Conclusions

- Better understanding of how the materials properties affect the thermal transport properties is need in order to develop thermal barrier materials
- Advanced experimental methodologies are needed for measurements of realistic sample configuration in order to capture the effects of features such as interfaces, surface roughness and density, which are missed during bulk measurements.
- Anisotropic thermal characterization was successfully achieved using TPS coupled with thermal imaging; and validating with a 3D modified Resistance in Series principle.
- The results showed that anisotropic thermal barriers can be used to create heat pathways to avoid overheating of electronics.
- Layers, composites and nanostructures are promising approaches to create anisotropic materials.

If successful, this research will address the need of high-thermal conductivity composite materials in emerging three-dimensional integrated circuit (3D IC), wide-bandgap semiconductor based power electronics, thermal barrier coatings (TBC) for engines and a variety of other technologies that needs smart thermal management solutions.

Acknowledgment

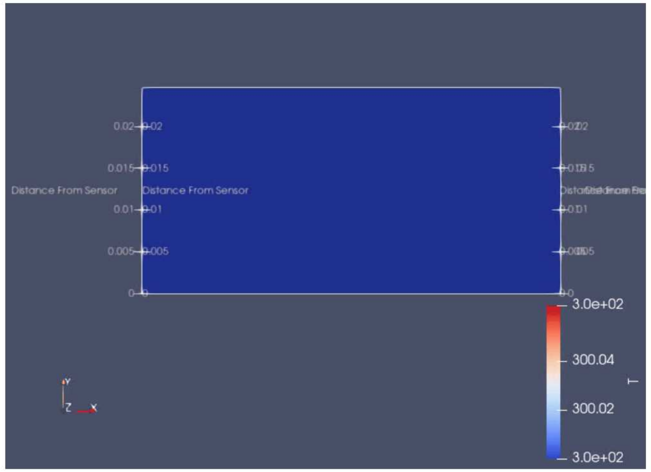
- Jacob Maher and Austin Acosta
- Matthew Walter and Anne Mallow
- Sandia Materials Chemistry Department

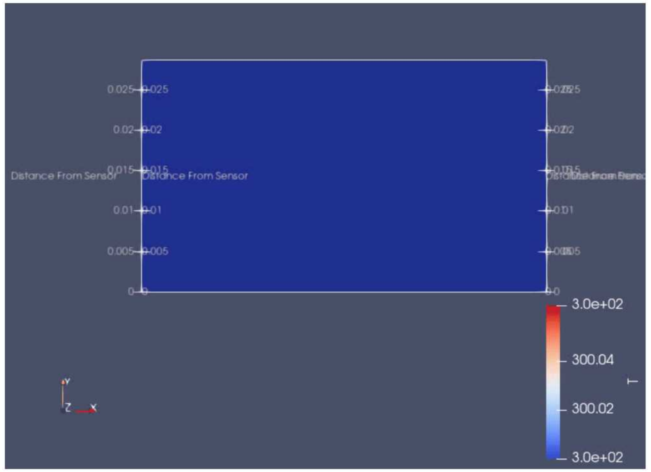
Videos

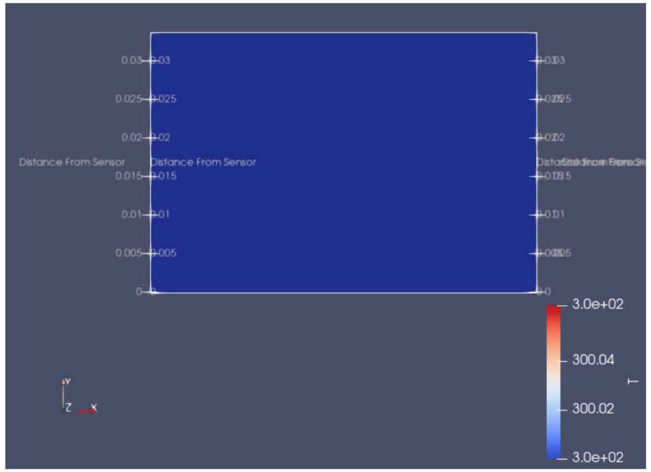
1mm Tflex

5mm Tflex

10mm Tflex







1mm Tflex

5mm Tflex

10mm Tflex

Click on videos