



Scalable Thermal Energy Technologies Seminar

Shannon Yee

Assistant Professor

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Scalable Thermal Energy Engineering Laboratory

Shannon Yee

- 2014 Assistant Professor
- 2013 PhD - Mech. Eng.
 - ✦ 2010 ARPA-E Fellow
 - ✦ 2008 Hertz Fellow
- 2008 MS - Nucl. Eng.
- 2007 BS - Mech. Eng.



Energy Club @ Georgia Tech
Heat Lab @ Georgia Tech
Grand Challenges @ Georgia Tech

Global Electrification



Fuel Resources



Global Cooling



Climate Change



Clean Water



Electric Transportation





Scalable *Thermal Energy Engineering* Laboratory

STEEL develops scalable thermal energy conversion & transport technologies to better utilize heat



“To Inspire, Educate, and Engage”

Seminar Topics:

(choose your own adventure)

Thermal Energy Conversion Technologies

1. [Thermoelectric Costs Scaling](#)
2. [Organic Thermoelectrics](#)
3. [Porous-Si Thermoelectric Cooling](#)
4. [Na Thermo-Electro-Chemical Heat Engine](#)
5. [Thermo-Electro-Chemical Cooling](#)
6. [Betavoltaic Energy Sources](#)

Thermal Transport Technologies

1. [Polymer Thermal Conductivity](#)
 2. [In-Plane FDTR Measurements](#)
 3. [Thermal Property Gas Sensing](#)
 4. [Better Building Materials](#)
- [Heat Lab @ Georgia Tech](#)
 - [Energy Club @ Georgia Tech](#)
 - [Grand Challenges @ Georgia Tech](#)

[New Directions with Organic Thermoelectrics](#)



New Directions with Organic Thermoelectrics

Prof. Shannon Yee

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Scalable Thermal Energy Engineering Laboratory

Georgia Institute of Technology

ORGANIC THERMOELECTRICS

Prof. Shannon Yee

Akanksha Menon – PhD Candidate

Rylan Wolfe – PhD Candidate

Sampath Kommandur – PhD Candidate

Dr. Kiarash Gordiz – Colorado School of Mines

Dr. Hend Elmoghunni – Qatar STP

Arnold Eng – PNNL

Olivia Meek – Landis & Gyr

Prof. John Reynolds

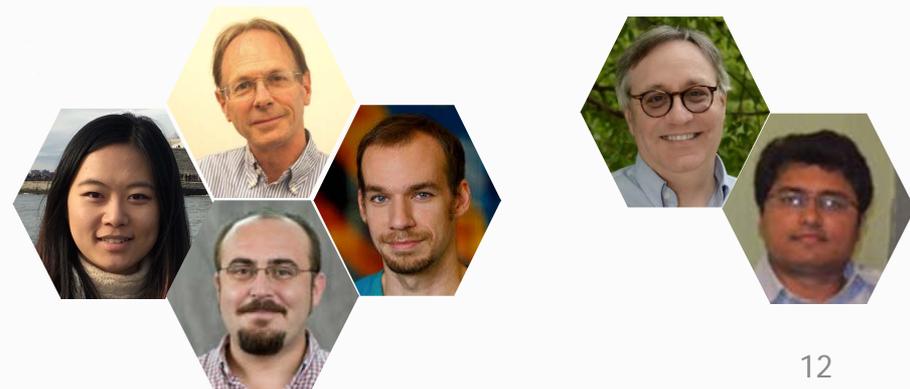
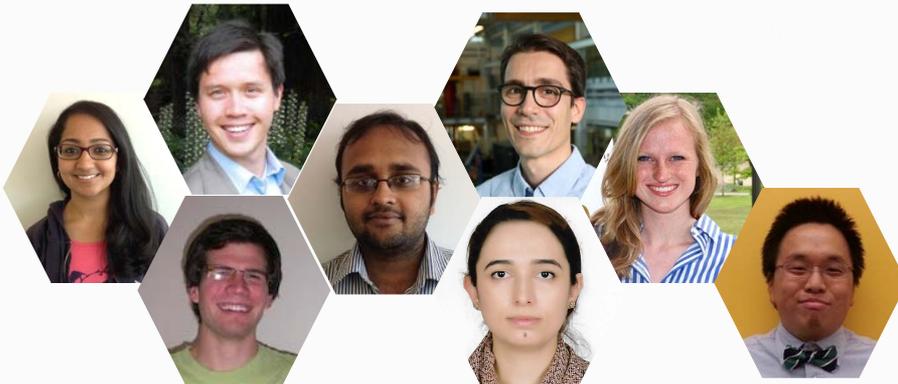
Dr. Bing Xu

Dr. Urdal Uzunlar

Dr. James Ponder – Imperial College

Prof. Seth Marder

Dr. Desari “Reddy” Raghunath



Part I

THERMOELECTRIC TECHNO-ECONOMICS

Thermoelectric Figure(s) of Merit

Power Generation



$\$/W$

Electric Cooling



$\$/kWh_{th}$

Cost {\$} - *Material, Manufacturing, & BOS*

- *Raw material costs*
- *Manufacturing costs*
- *Balance of System (BOS) costs*
 - *Heat Exchangers*
 - *Insulating Plates*
 - *Metallization*

$$G = \frac{C}{P}$$

Power {W} - *Device Physics*

“A \$/W METRIC FOR THERMOELECTRIC POWER GENERATION: BEYOND ZT”

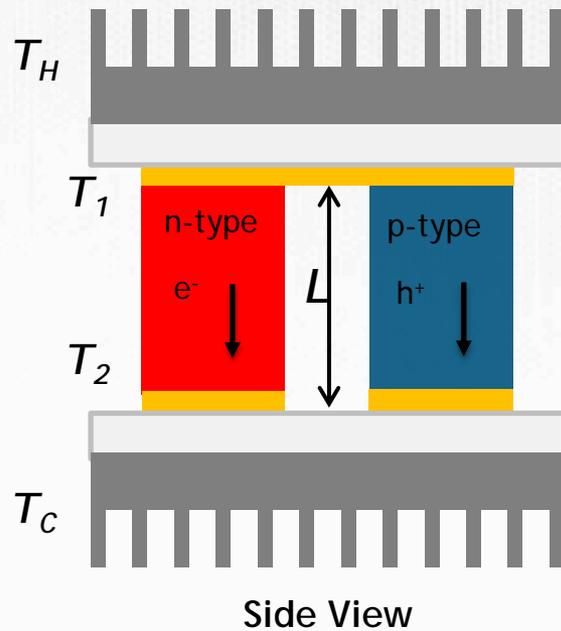
S. Yee *et al.*, *Energy & Environ. Sci.*, **6**, 2561-2571, 2013.

S. LeBlanc, S. Yee, *et al.*, *Ren. Sus. Energy Rev.*, **32**, 313-327, 2014.

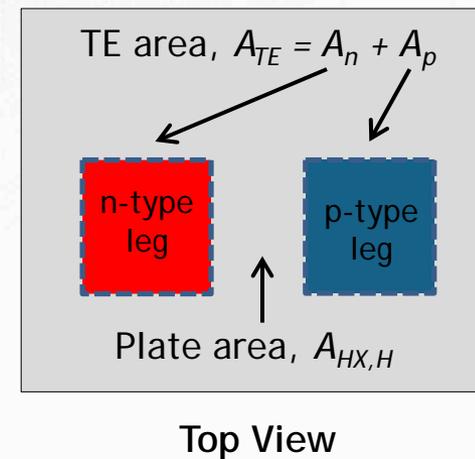
T. Hendricks, S. Yee, S. LeBlanc, *J. Electron. Mat.* 2015.

Thermoelectric Device Topology

Design variables - leg length, L & fill factor, F



$$F = \frac{A_{TE}}{A_{HX,H}}$$



Costs: Material, Manufacturing, & Heat Exchangers Costs

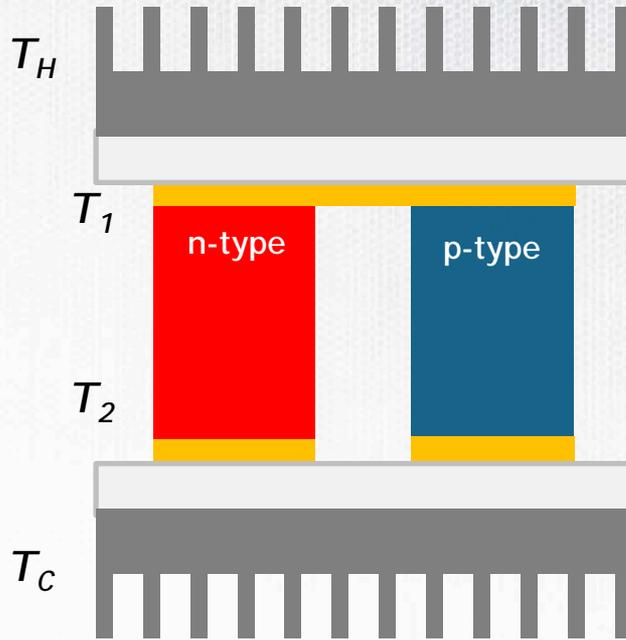
- Volumetric Costs, C''' [\$/m³]
 - Material costs
 - Manufacturing costs (dicing, cutting, *etc.*)
- Areal Module Costs, C'' [\$/m²]
 - Metallization, metal shunts, *etc.*
 - Manufacturing costs (depositions, assembly, *etc.*)
- Heat Exchanger Costs, C_{HX} [\$/ (W/K)]
 - Ceramic plates, plate-and-fin, *etc.*

$$C = (C'''L + C'')AF + C_{HX}UA$$

[$\$$] [\$/m³] [\$/m²] [\$/ (W/K)]

Power: Thermoelectric Device

Physics

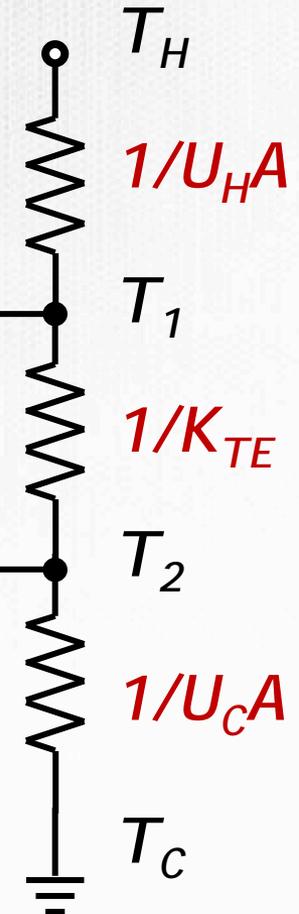


Side View

$$I^2 R_{el} / 2 - I S_{pn} T_1$$

$$1/K_{||}$$

$$I^2 R_{el} / 2 + I S_{pn} T_2$$



$$P = \frac{S_{pn}^2 (T_1 - T_2)^2}{R_{el}} \left(\frac{m}{(m+1)^2} \right)$$

$$m = \frac{R_L}{R_{el}}$$

Dimensional and Non-dimensional Terms

Length [m]

$$L_T \equiv \frac{k}{U}$$

Thermal Length

$$L_C \equiv \frac{C''}{C'''}$$

Cost Length

$$L_{HX} \equiv \frac{C_{HX} U}{C'''}$$

Heat Exchanger Length

Dimensionless

$$\tilde{L} \equiv \frac{L}{L_T}$$

$$\tilde{L}_C \equiv \frac{L_C}{L_T}$$

$$\tilde{L}_{HX} \equiv \frac{L_{HX}}{L_T}$$

Cost Metric for Thermoelectrics

Load Matching

$$\frac{G}{G_0} \approx \frac{1}{4} \left(\frac{(m+1)^2}{m} \right) (2F + \tilde{L})^2 \left(1 + \frac{\tilde{L}_C}{\tilde{L}} + \frac{\tilde{L}_{HX}}{\tilde{L}F} \right)$$

Three Cost Terms

Fill Factor
Leg Length

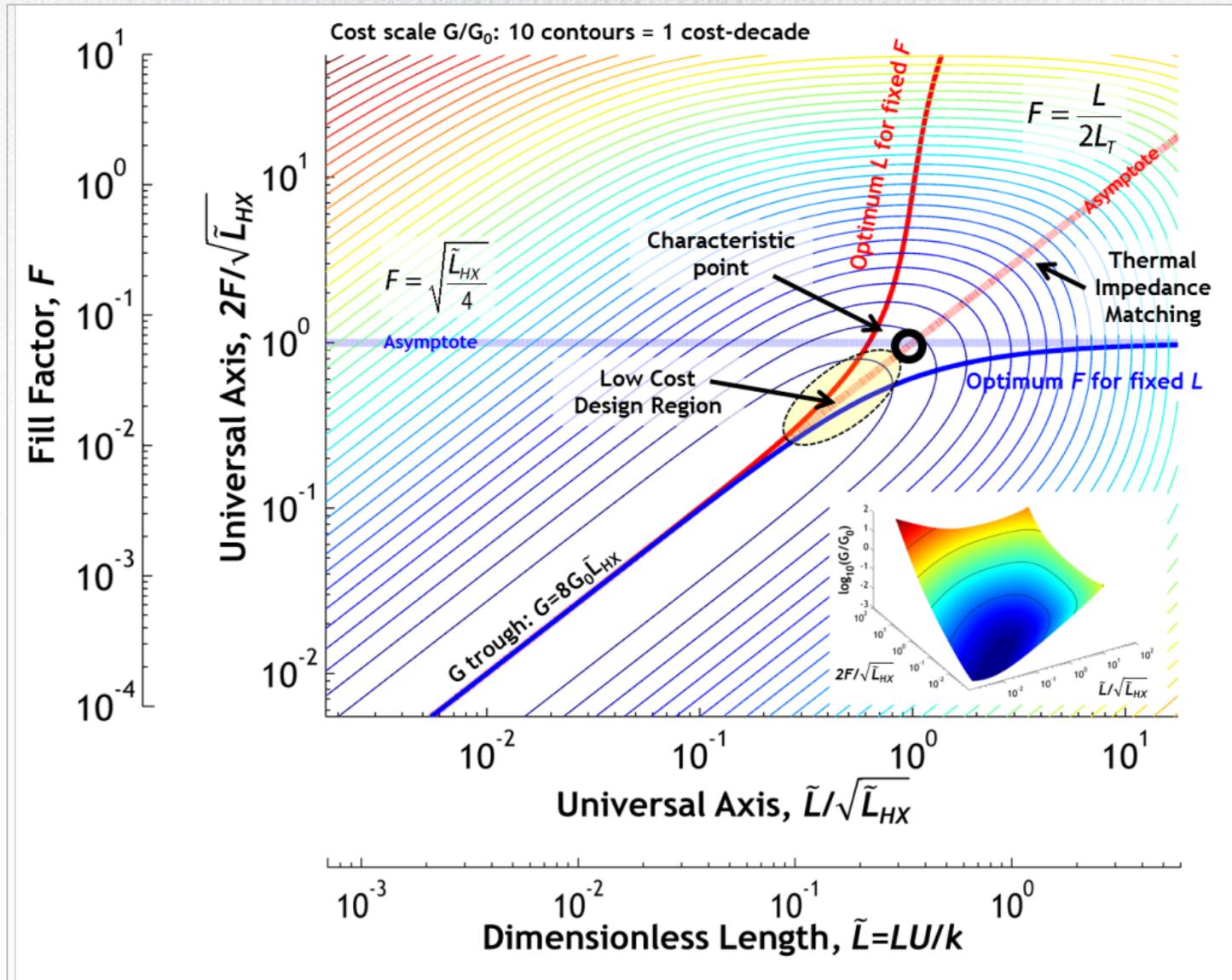
Volumetric Cost [\$/m³]

Volume [m³]

$$G_0 = \frac{C''' L_T A}{\frac{1}{4} Z T_H \left(\frac{T_H - T_C}{T_H} \right) UA (T_H - T_C)}$$

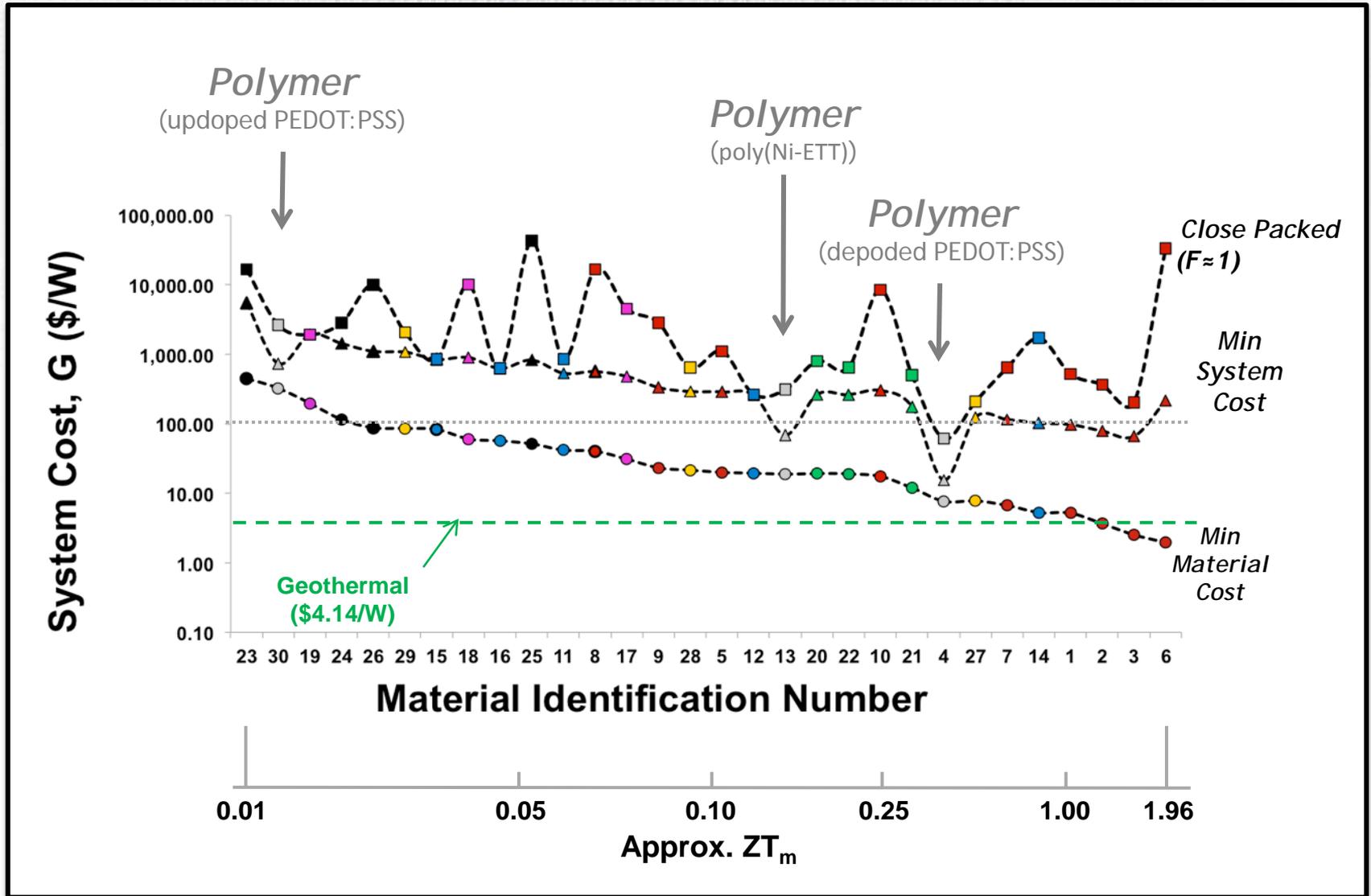
small ZT efficiency
Carnot efficiency
Thermal potential [W]

Design Space for G in \$/W

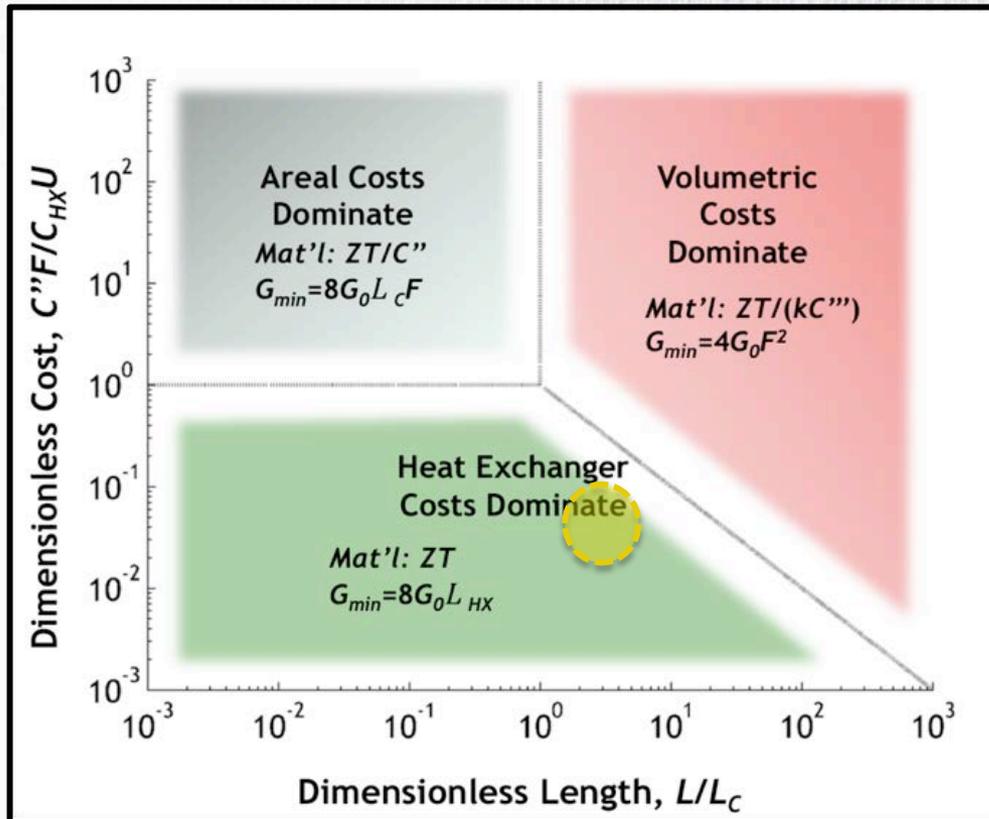


\$/W Cost Comparison: Low Temp Waste Heat

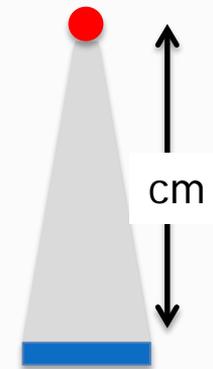
$$T_H = 100^\circ\text{C}, T_C = 20^\circ\text{C}$$



\$/W Cost Minimized Design



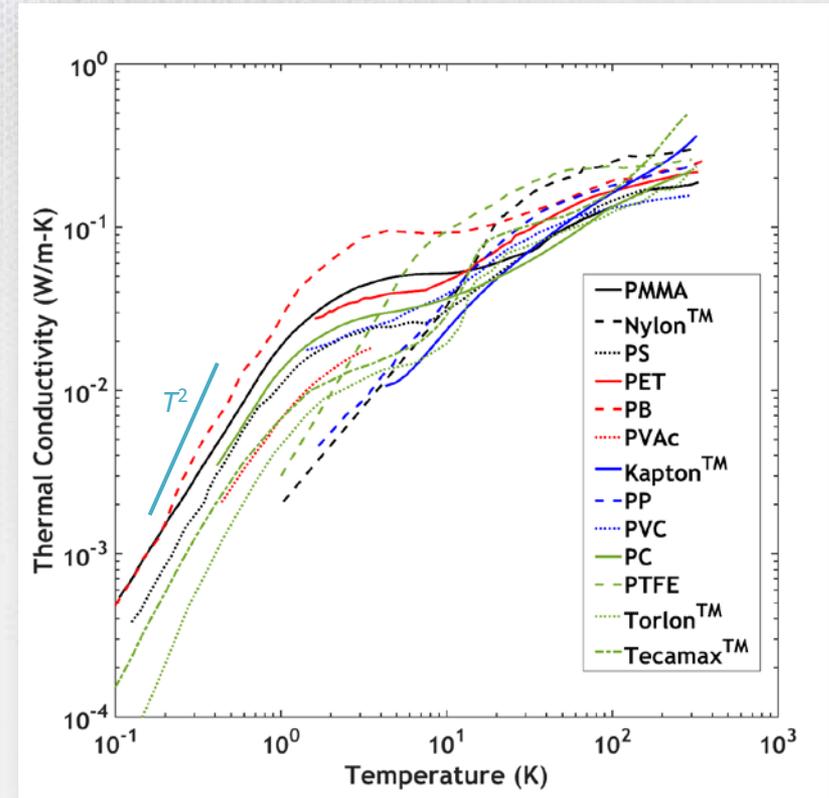
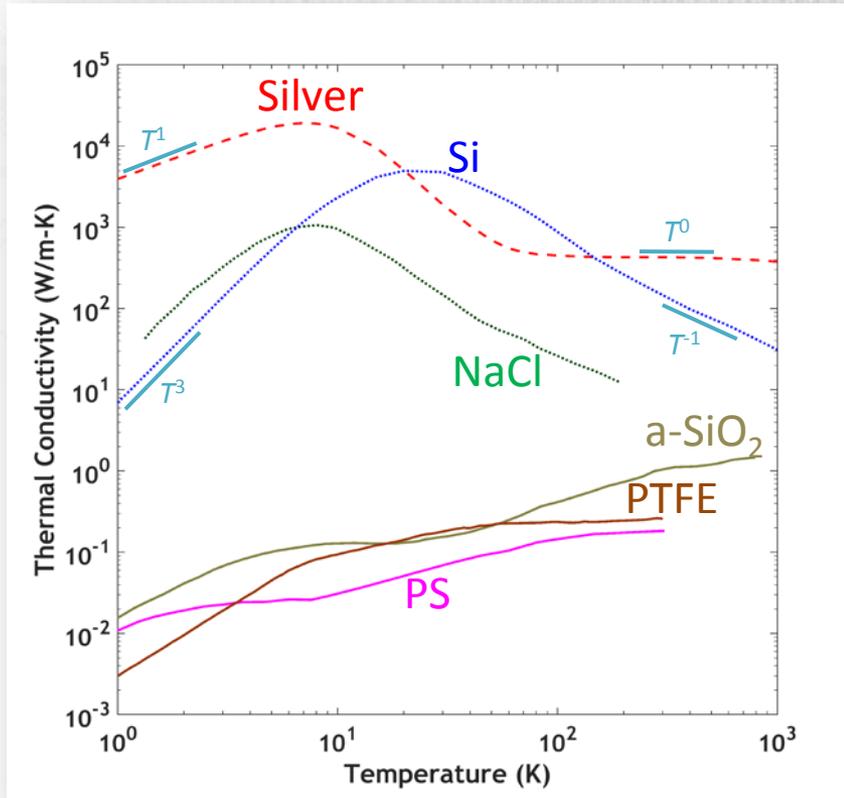
- Heat exchanger costs dominate have dominated in ALL systems to-date
- Why not eliminate the heat exchanger and operate with natural convection?
 - $k_{eff} < 0.5 \text{ W/m-K}$
 - $L_{opt} \sim 5 \text{ cm}$
 - $F_{opt} > 1$



Part II

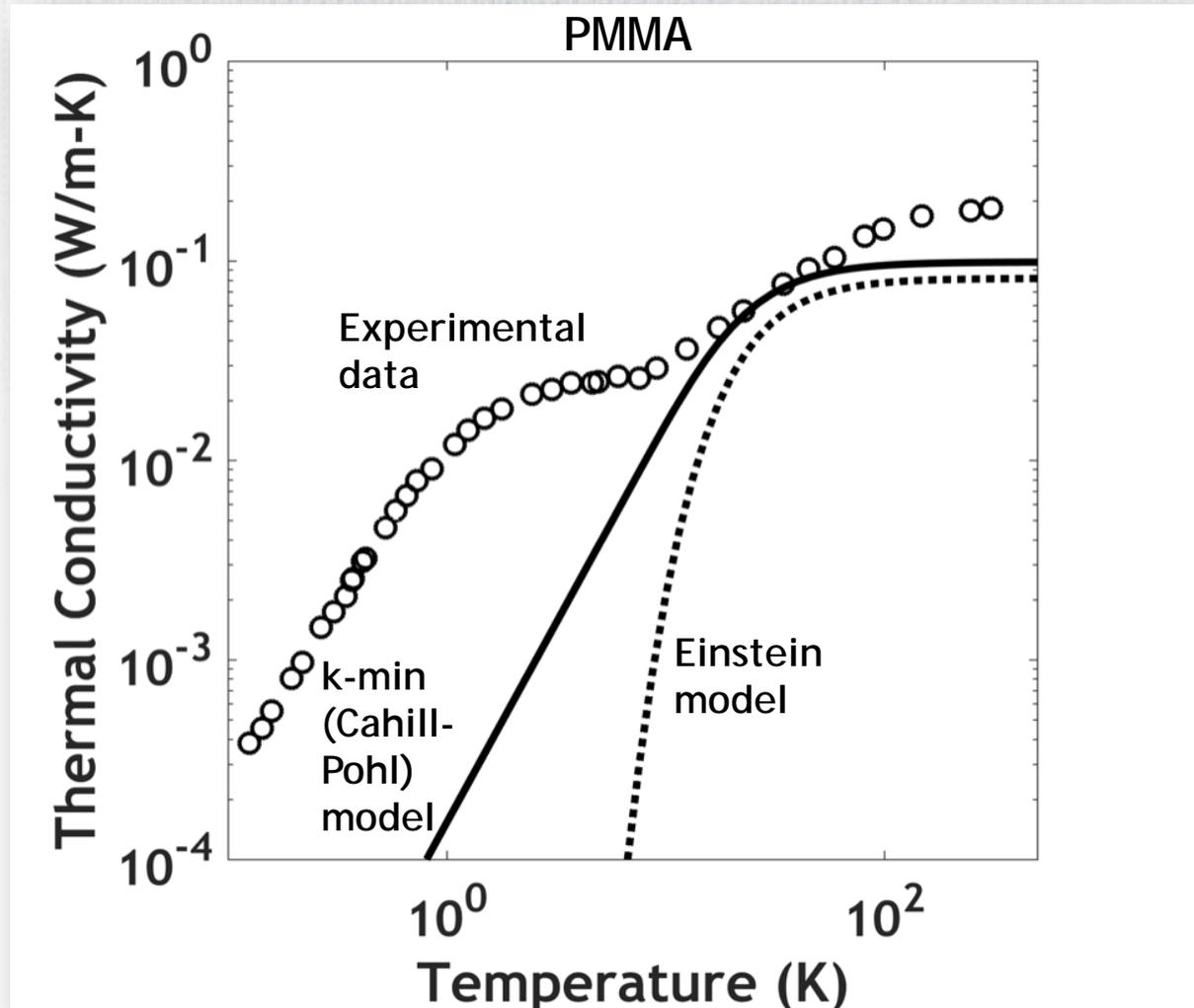
THERMAL TRANSPORT IN POLYMERS

Experimental Observations of Thermal Conductivity

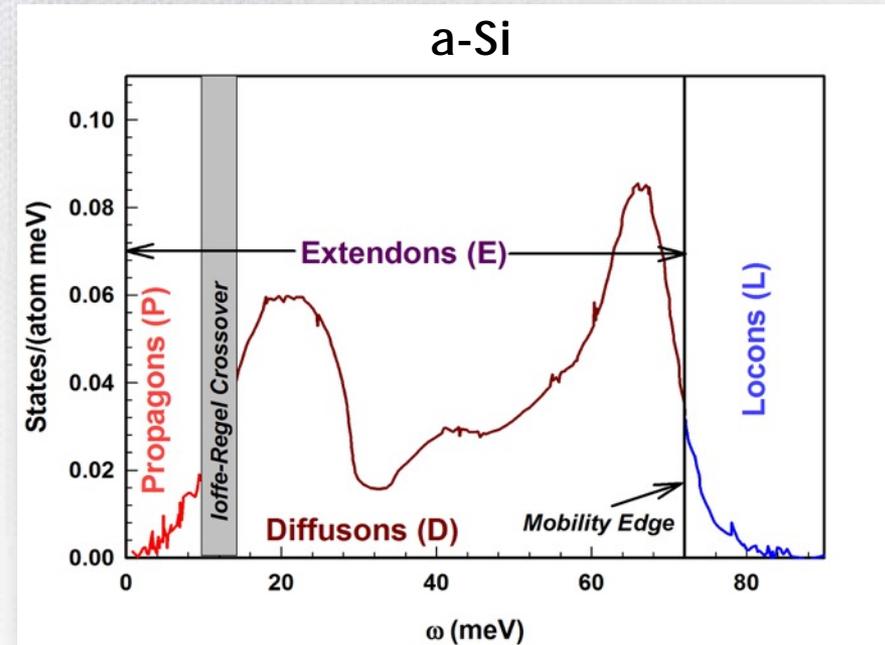
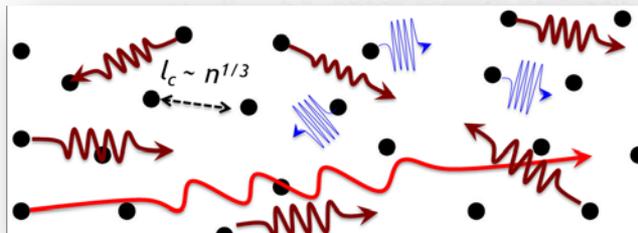


Amorphous polymers exhibit (i) monotonically increasing thermal conductivity trends and (ii) undergo plateau-like transition(s)

Comparison of experimental data to amorphous thermal conductivity models



Vibrons are vibrational modes in amorphous materials

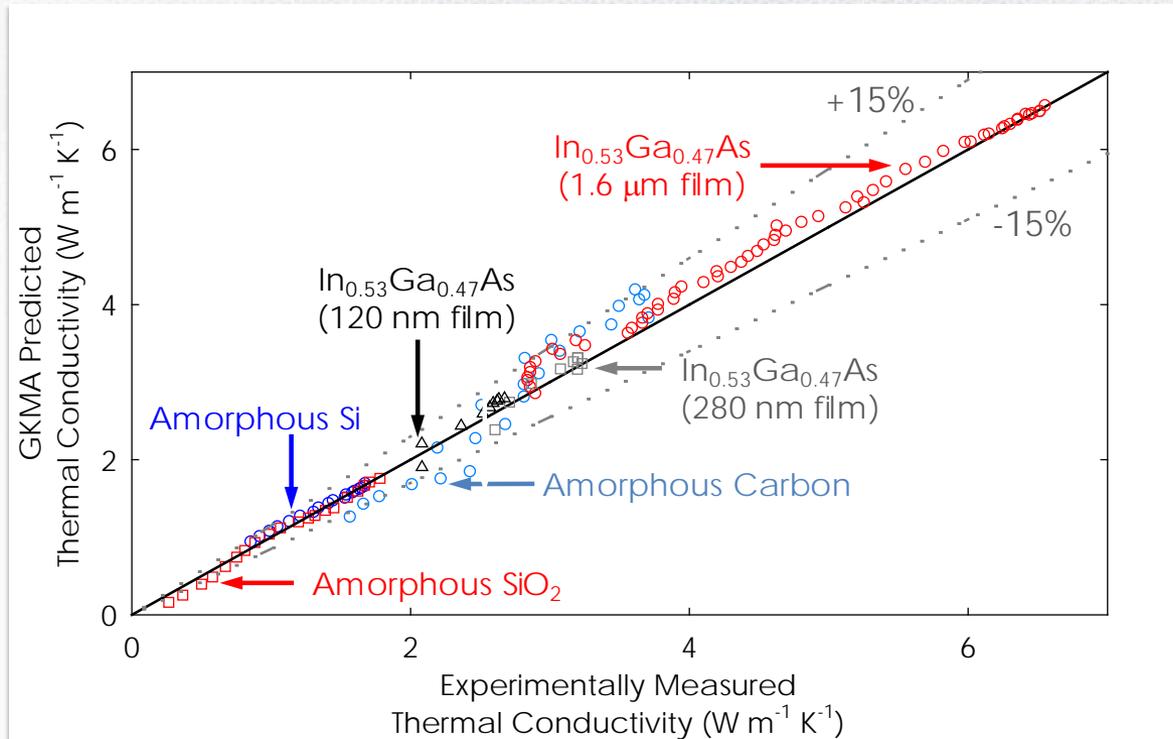


$$l_{mfp} > a \quad l_{mfp} \sim a \quad l_{mfp} < a \quad l_{mfp} \ll a$$

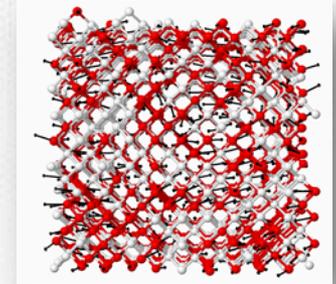
The concepts of a mean free path and a wave-vector are less useful for diffusons and locons.

Green-Kubo Modal Analysis (GKMA)

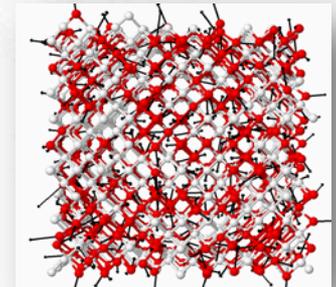
Uses super-cell lattice dynamics (SCLD) with molecular dynamics (MD) to calculate the contributions of phonons in a general way. Can be applied to amorphous materials, polymers, crystals alloys etc. using one unified formalism. When disorder exists, phonons no longer look like waves.



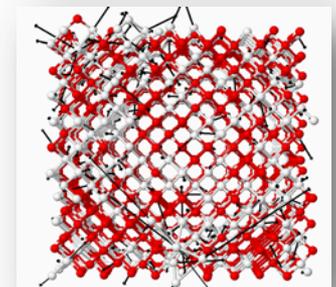
Propagons



Diffusons



Locons



W. Lv, A. Henry, Direct Calculation of Modal Contributions to Thermal Conductivity via Green-Kubo Modal Analysis: Crystalline and Amorphous Silicon, *New Journal of Physics*, 18, 013028 (2016).

Seyf, Hamid Reza, et al. Rethinking phonons: The issue of disorder, *npj Computational Materials* 3, 1, 49 (2017)

K. Gordiz and A. Henry, A formalism for calculating the modal contributions to thermal interface conductance, *New Journal of Physics*, 17, 103002 (2015)

Origins of Polymer Thermal Conductivity

$$k = \frac{1}{3} cvl \quad \xrightarrow[\text{Diffusivity}]{D = vl} \quad k = \frac{1}{3} cD$$

- Independent contributions from each vibron

$$k = k_P + k_D + k_L$$

$$k = \frac{1}{3} \sum_i \int_{\omega_{i-1}}^{\omega_i} c(\omega) D(\omega) N(\omega) d\omega$$

Vol. Specific heat

$$c = 3nk_B \frac{x^2 e^x}{(e^x - 1)^2}$$

Density of states

$$N_i(\omega) \propto \frac{3\omega^2}{2n\pi^2 v_s^3}$$

Diffusivity

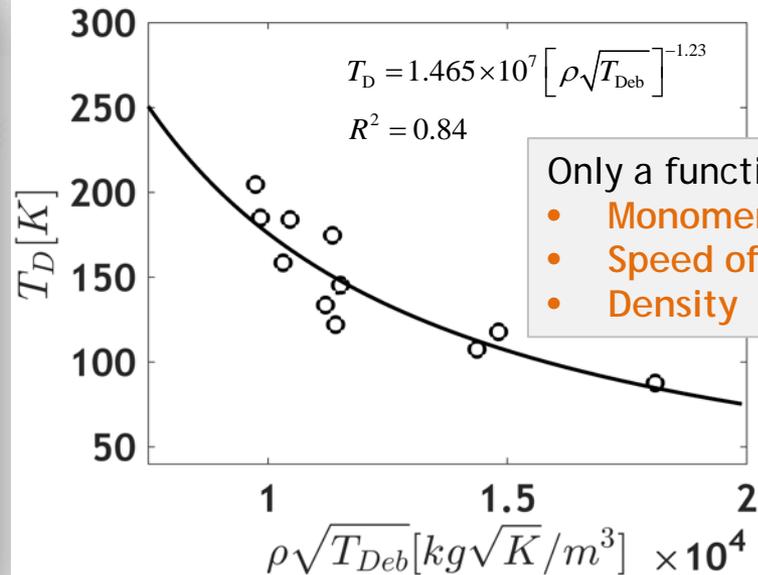
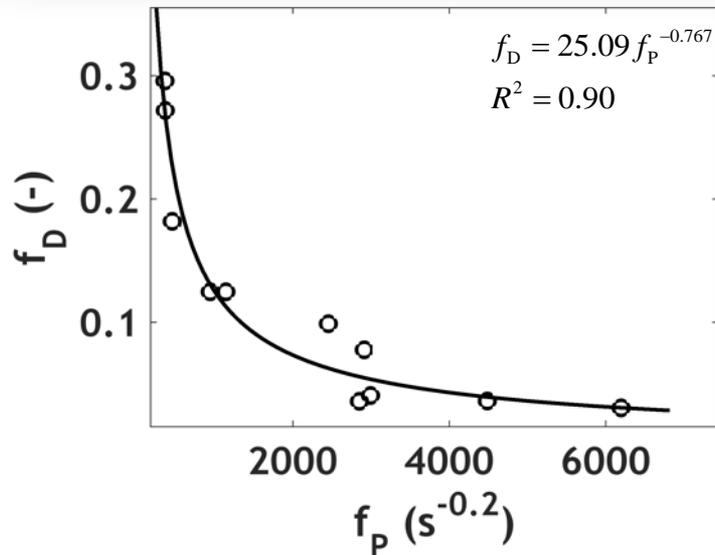
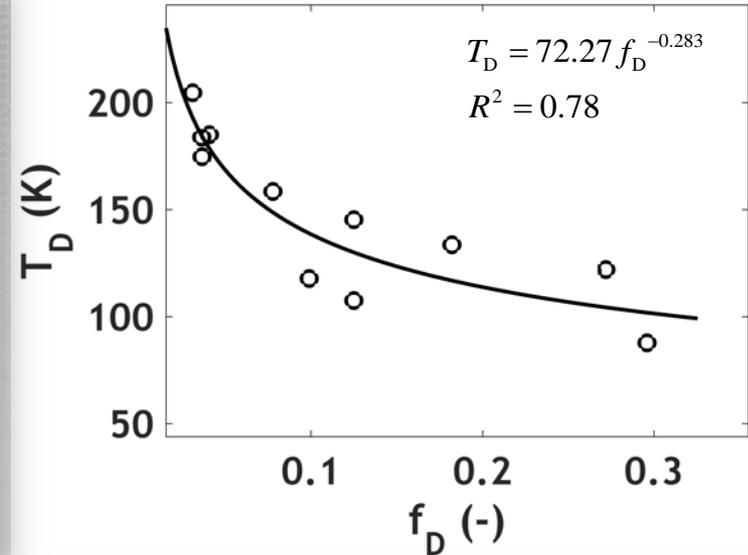
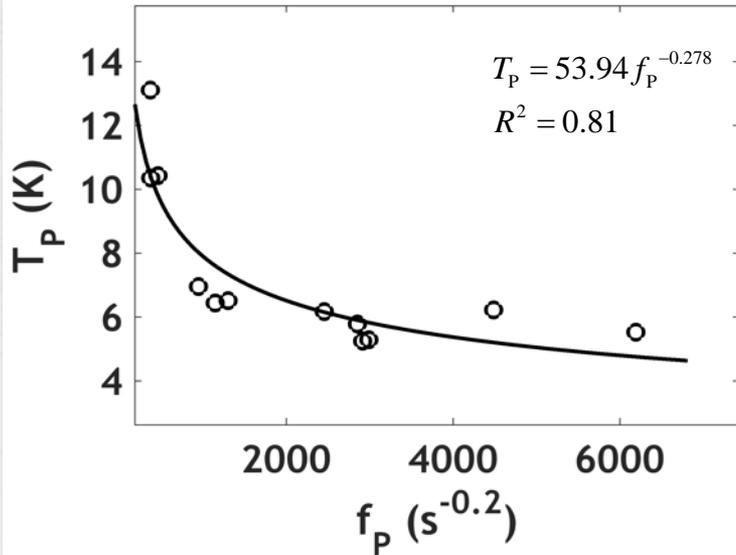
$$D_i(\omega) \propto \omega^{-\beta_i}$$

Simplified Model

$$k(T) = \underbrace{f_P \frac{k_B}{v_s} \left(\frac{k_B T}{\hbar} \right)^{1.8} \int_0^{x_P} \frac{x^{2.8} e^x}{(e^x - 1)^2} dx}_{\text{Propagons}} + \underbrace{f_D \frac{k_B}{v_s} \left(\frac{k_B T}{\hbar} \right)^2 \int_{x_P}^{x_D} \frac{x^3 e^x}{(e^x - 1)^2} dx}_{\text{Diffusons}}$$

- **Considers only propagons and diffusons**
 - Neglect locons because polymers degrade/melt/glass-transition as locons contribution becomes significant
- **Fixed value of β in the diffusivity relation**
 - $T^{1.8}$ has good agreement with experimental data for low T
 - T^2 has good agreement with experimental data for mid T
- **Reduces to 4 parameters (T_P, f_P, T_D, f_D)**

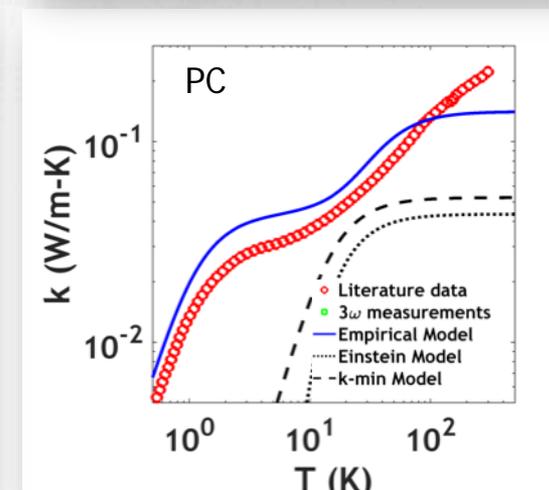
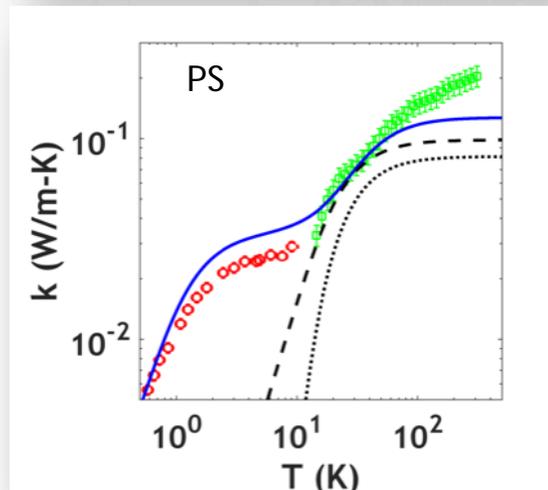
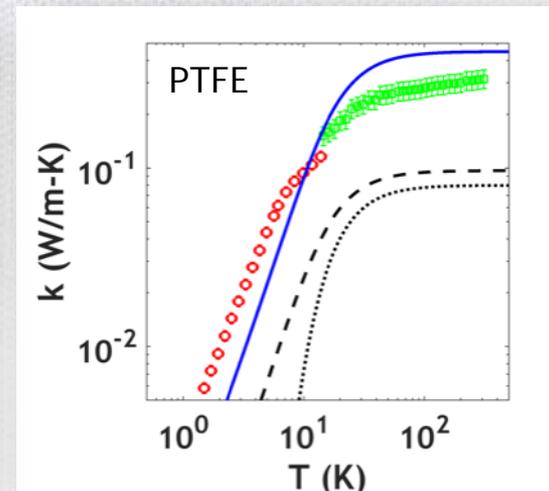
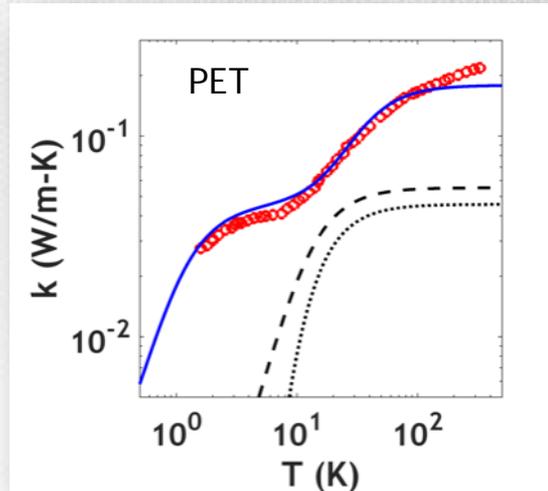
Empirical Observations Relating Parameters



Only a function of

- Monomer Weight
- Speed of Sound
- Density

Empirical model predicts the temperature dependent thermal conductivity well



Interesting Model Interpretations

- MFP cross-over between propagons and diffusons

$$\frac{l_{\text{mfp}}}{a} \approx \frac{2\pi v_s n^{1/3}}{\omega_p} \approx 10$$

- Modal contribution to thermal conductivity

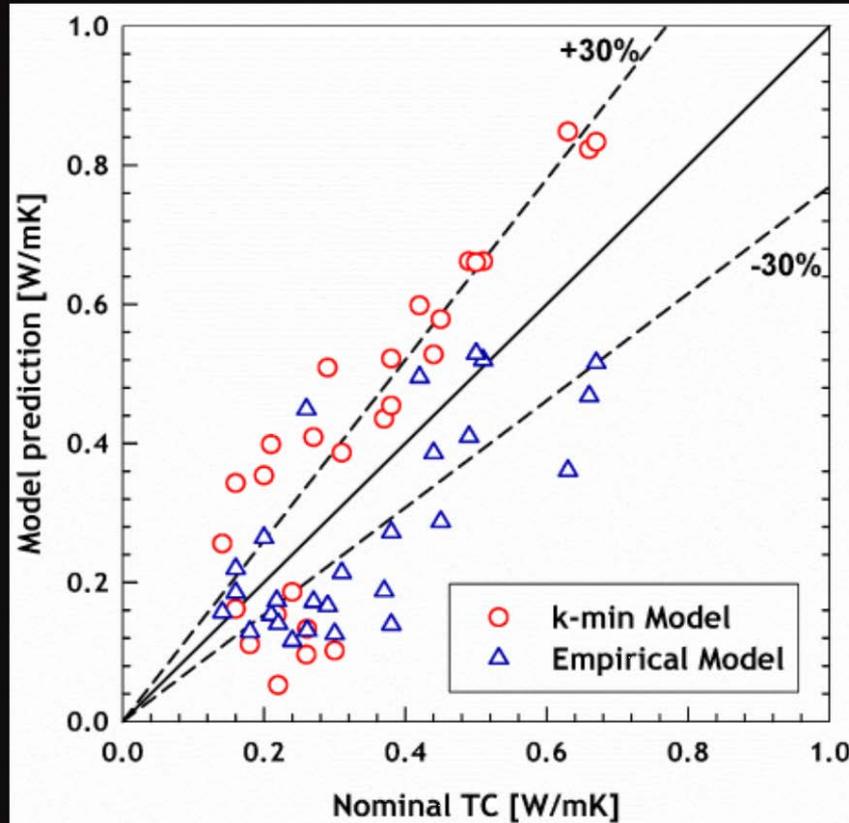
$$k_p(T) = 1.85 \frac{f_p^{0.5}}{v_s} \left[\frac{1}{x_p^{1.8}} \int_0^{x_p} \frac{x^{2.8} e^x}{(e^x - 1)^2} dx \right]$$

$$k_D(T) = 1226 \frac{f_D^{0.43}}{v_s} \left[\frac{1}{x_D^2} \int_{x_p}^{x_D} \frac{x^3 e^x}{(e^x - 1)^2} dx \right]$$

- Prediction of room temp. thermal conductivity for amorphous polymers

$$k_{\text{max}} = \frac{1}{v_s} \left(f_p^{0.5} + 613 f_D^{0.43} \right)$$

Room Temperature Amorphous Polymer Thermal Conductivity



An upper limit to the thermal conductivity in amorphous polymers

$$k_{\max} = \frac{1}{v_s} \left(f_P^{0.5} + 613 f_D^{0.43} \right)$$

$$f_P \propto \rho^{-0.3} v_s^{-1.7} M^{0.55}$$

$$f_D \propto \rho^{4.3} v_s^{2.2} M^{-0.7}$$

ρ	Density (kg/m ³)
M	Monomer molar mass (kg/mol)
v_s	Speed of sound* (m/s)

* averaged over the two modes

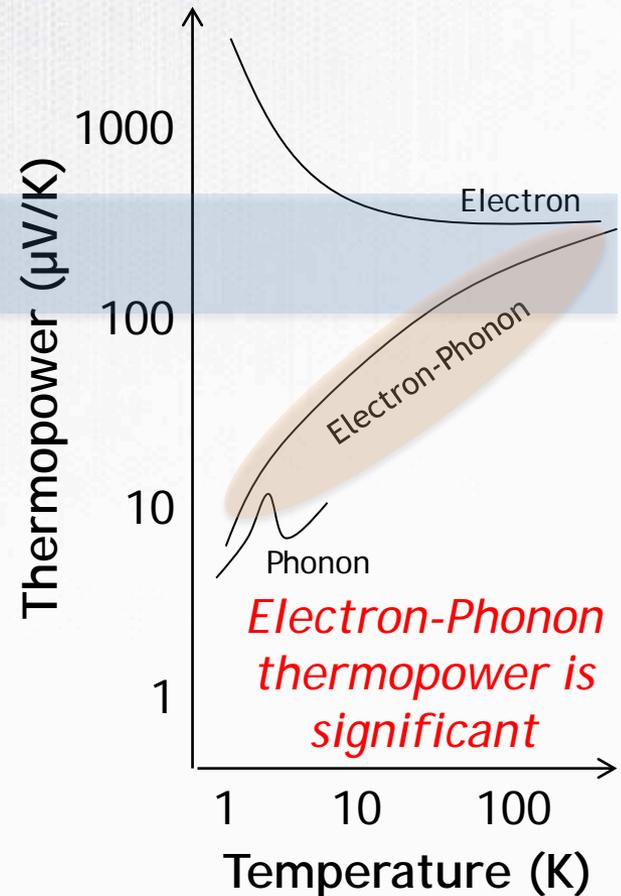
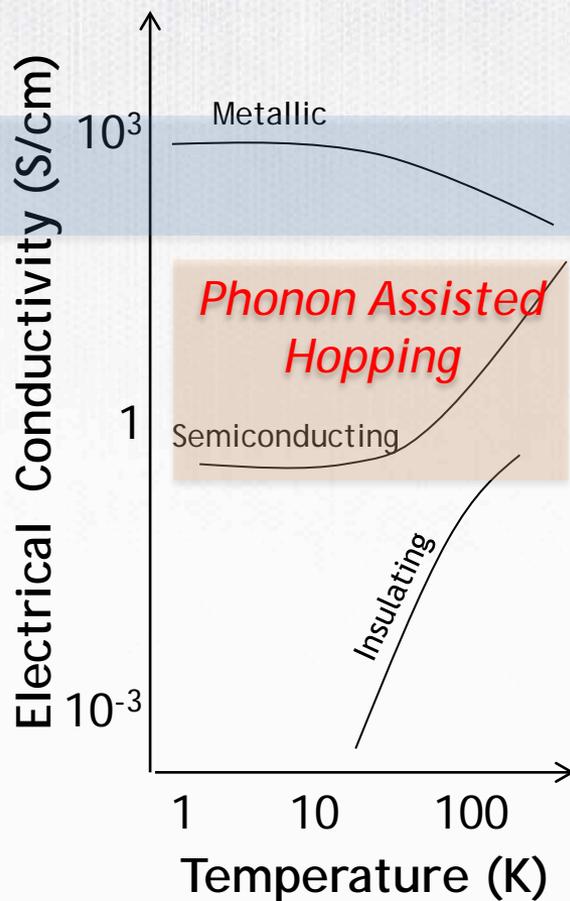
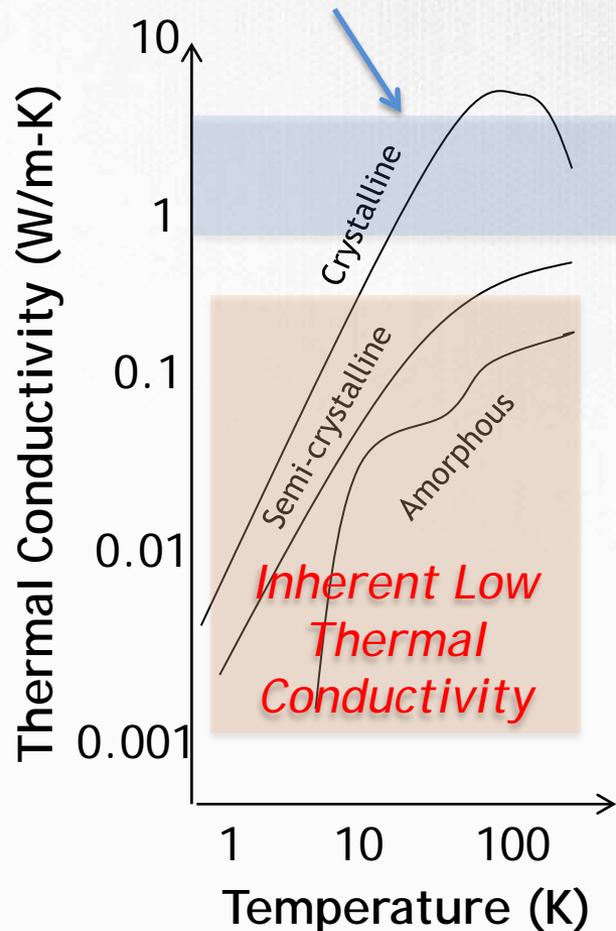
S. Kommandur and S.K. Yee, *J. Poly. Sci. B*, 55, 2017

X. Xie et al., *Macromolecules*, 49, 2016

X. Xie et al., *Phys. Rev. B*, 95, 2017

OTEs offer new directions for improving TE transport

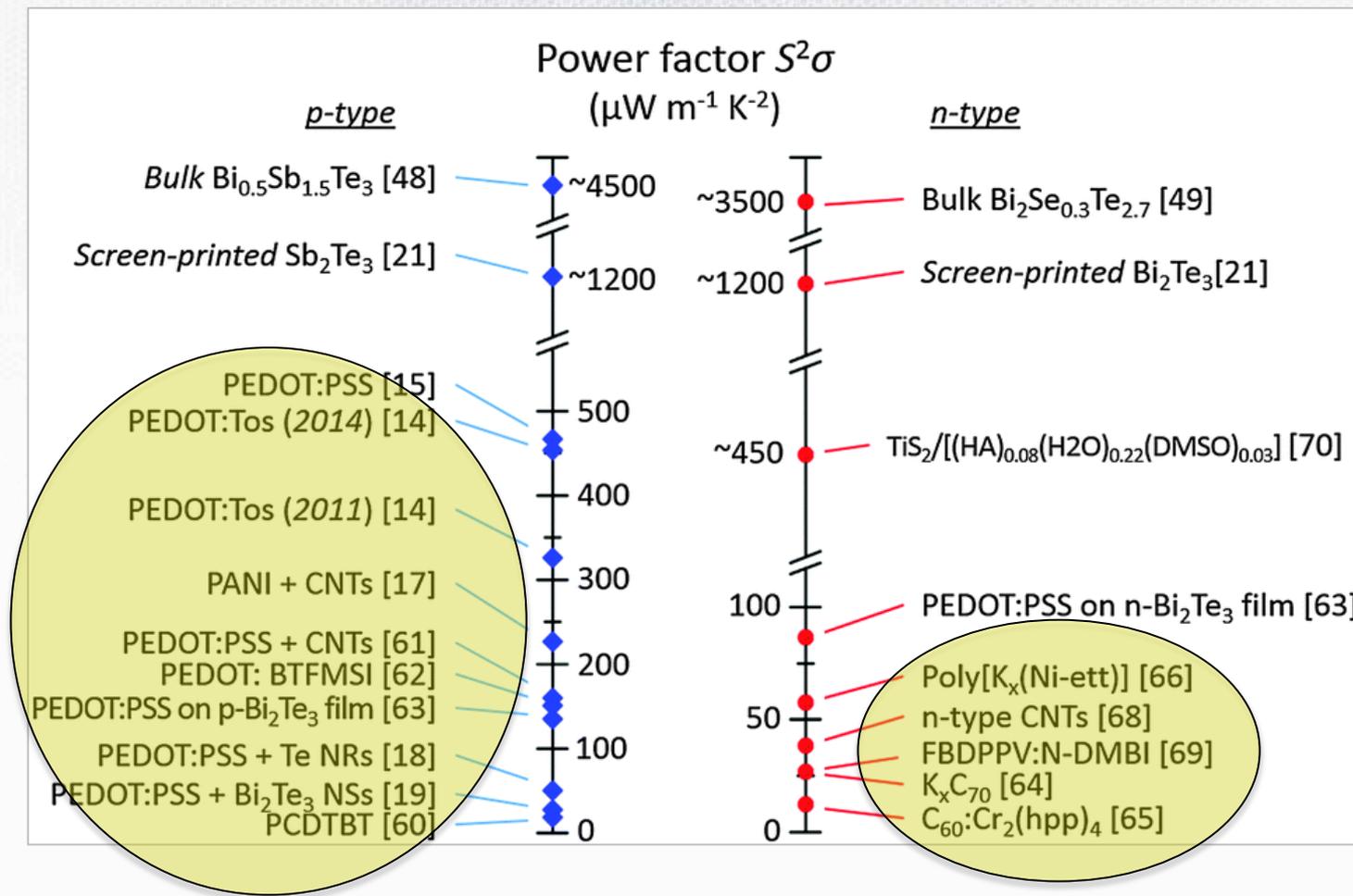
Most conventional Inorganic TEs



Part III

POLYMER THERMOELECTRIC MATERIALS

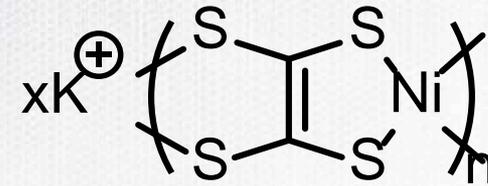
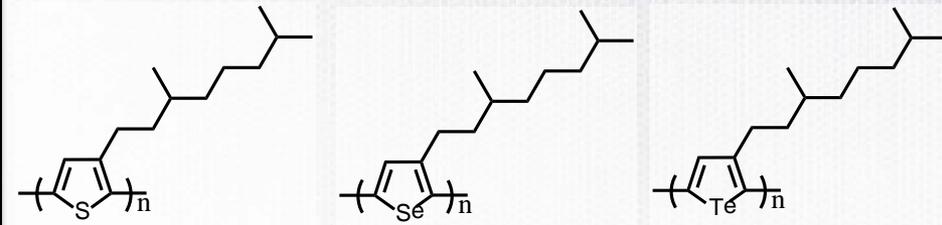
State-of-the-Art Flexible TEs



Thermoelectric Polymers

p-type

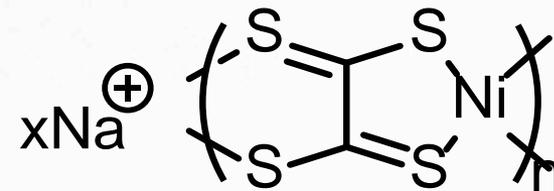
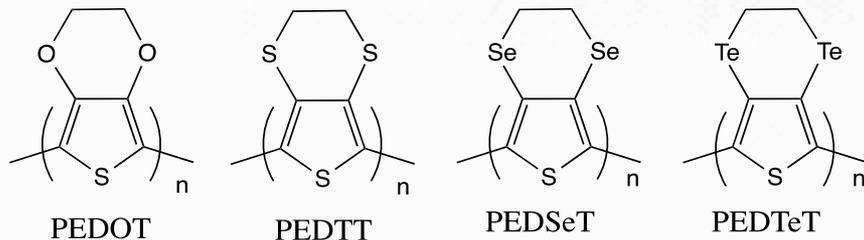
n-type



Poly(Ni-ethenetetrathiolate)

Ni-ETT

*In collaboration with Prof. Dwight Seferos
University of Toronto*

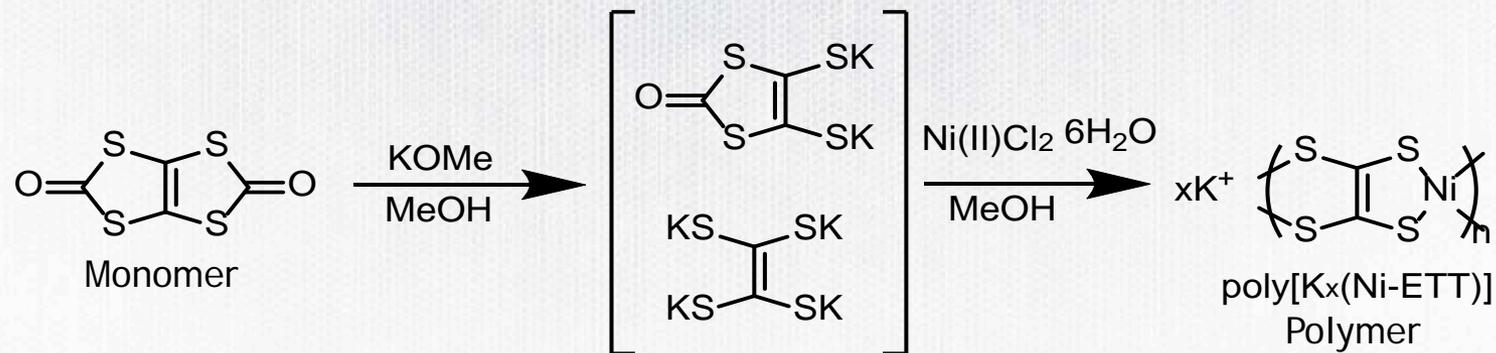


Poly(Ni-tetrathiooxalate)

Ni-TTO

*In collaboration with Drs. Peter Bonneson & Kulun Hong
Oak Ridge National Laboratory*

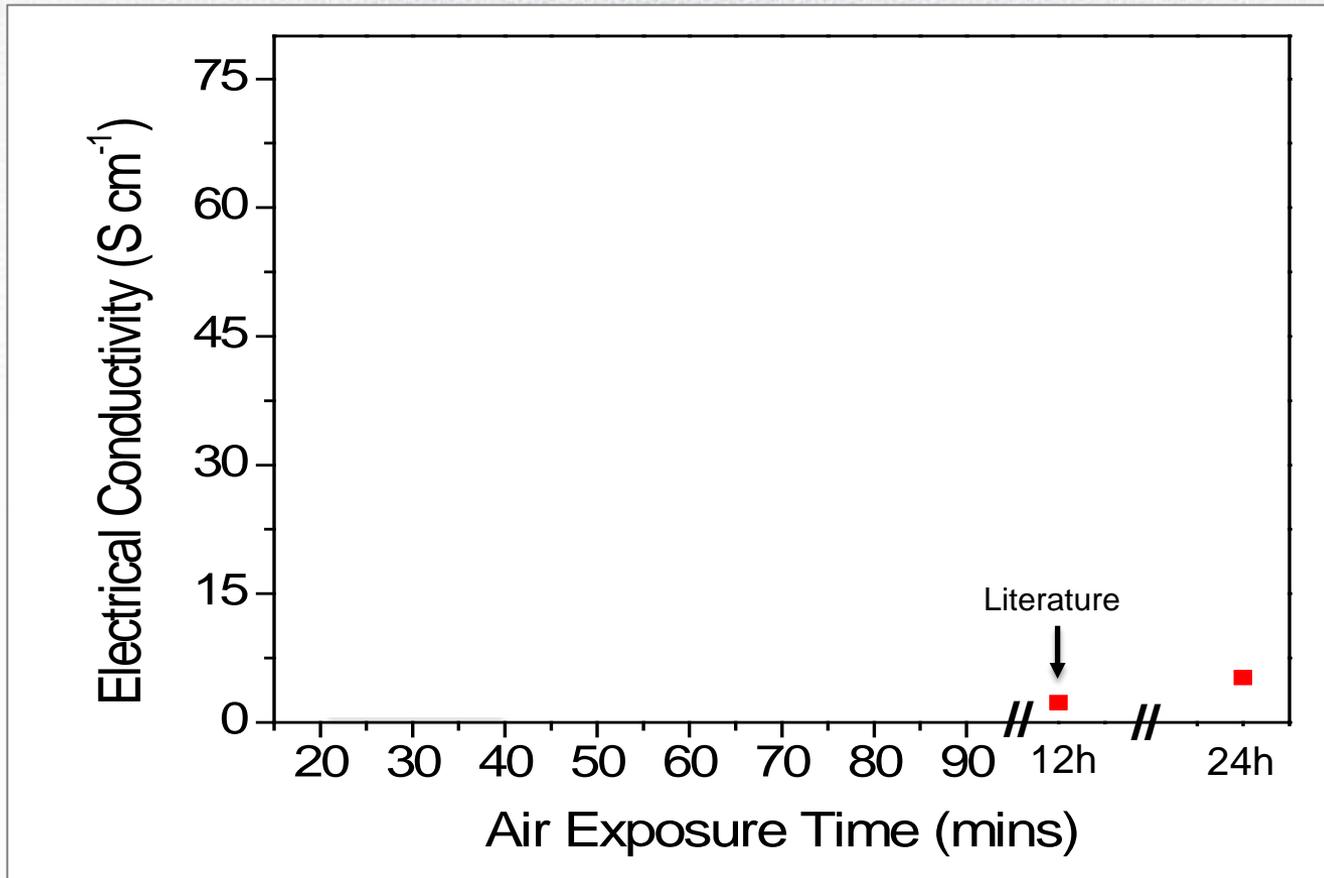
Synthesis of Ni-ETT and Varying the Oxidation Time



- (ir) is
- tion
- er is
- an in
- + DMS
- Limited characterization techniques
- Dissolve KOMe in methanol
- Add TPD monomer
- Add NiCl₂ and oxidize
- Filter and crush into powder

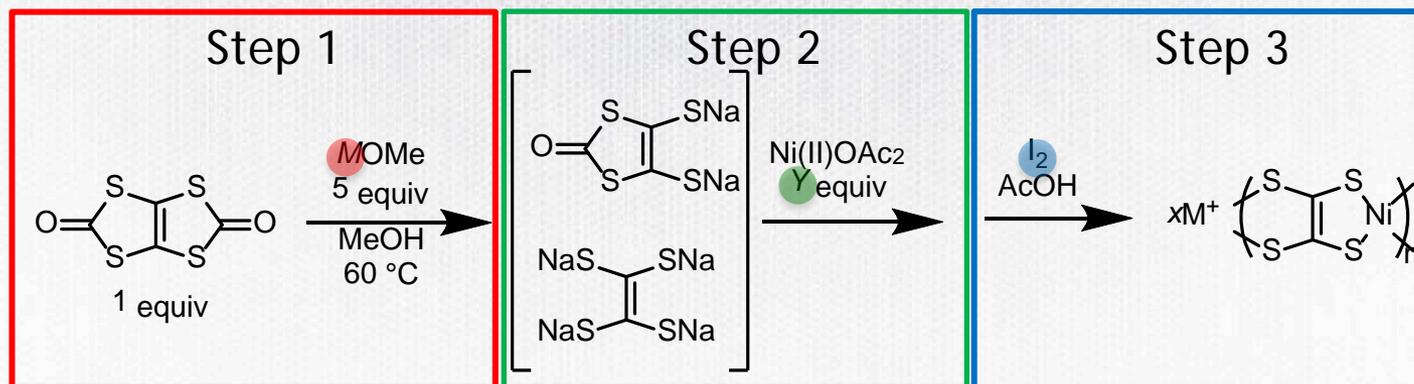


Tuning Electrical Conductivity via Oxidation

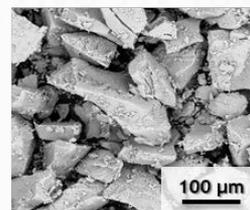
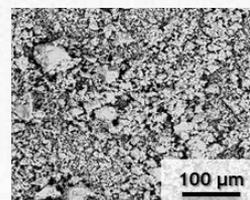


Seebeck coefficient $-30 \pm 3 \mu\text{V/K}$ and thermal conductivity is $0.27 \pm 0.02 \text{ W/m-K}$ remain unchanged

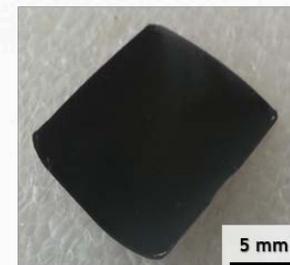
Synthesis of ETT and Film Post-Treatment Allows for Tuning Properties



Sol-gel



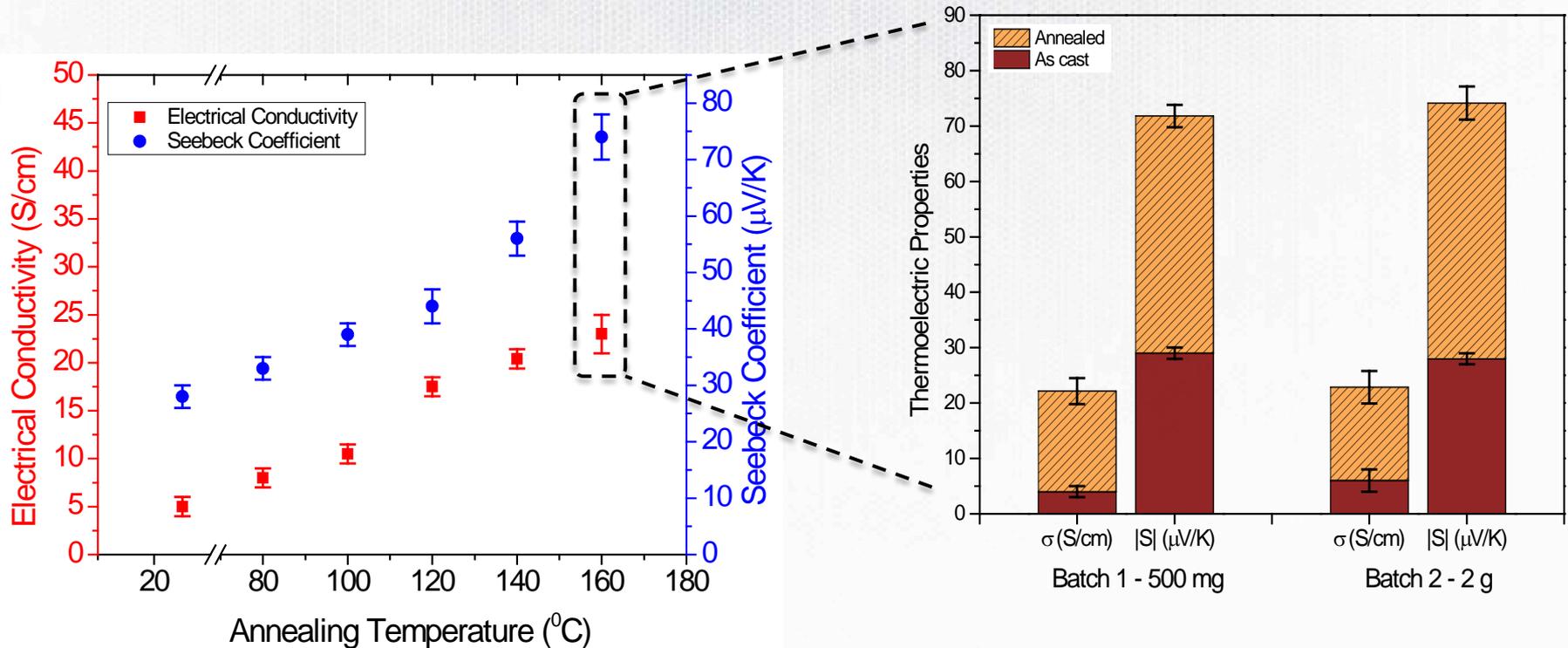
SEM of NiETT



NiETT film with PVDF/DMSO

Measurements at 25 °C with 4:1 w/w ETT:PVDF/DMSO (matrix is 10 mg/mL).

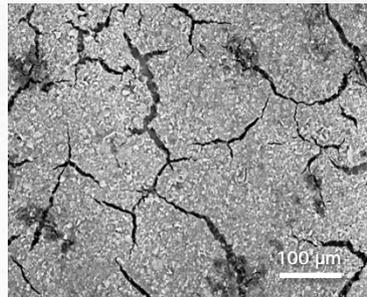
Film Post-Treatment by Annealing Simultaneously Improves S and σ



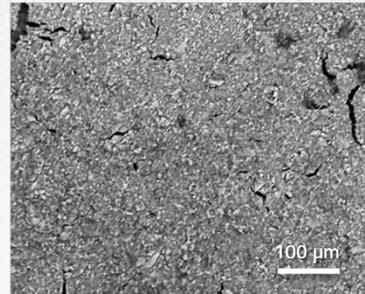
Annealing in air at 160°C for one hour enhances S and σ .
Annealing in air, nitrogen, and vacuum yield similar properties.

Morphological and Compositional Changes occur during Annealing

Morphological changes



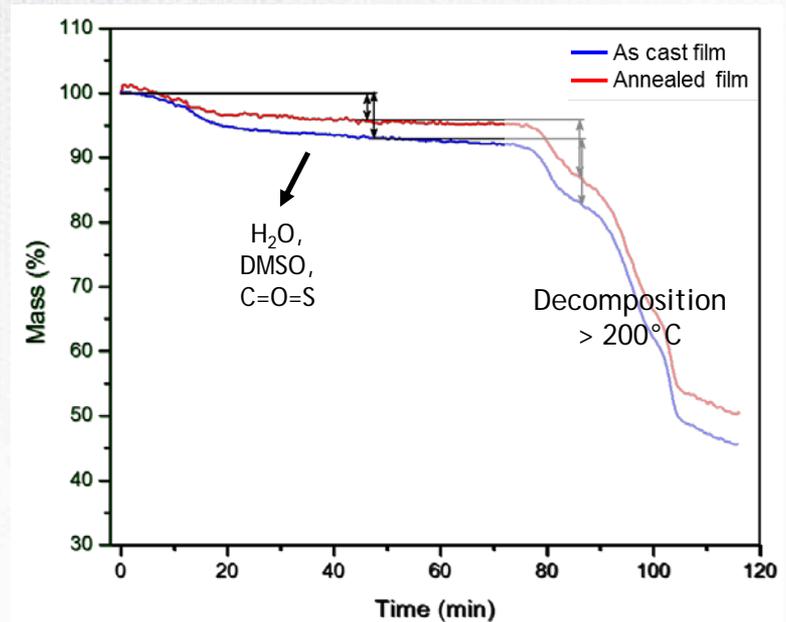
As cast film



Annealed film

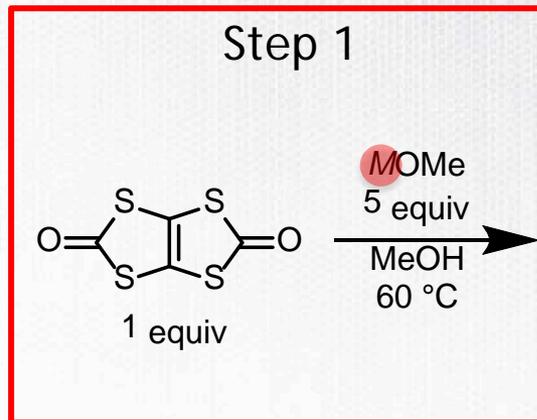
Annealing close to the melting temperature of PVDF improves network connectivity, resulting in a higher σ value.

Chemical composition changes

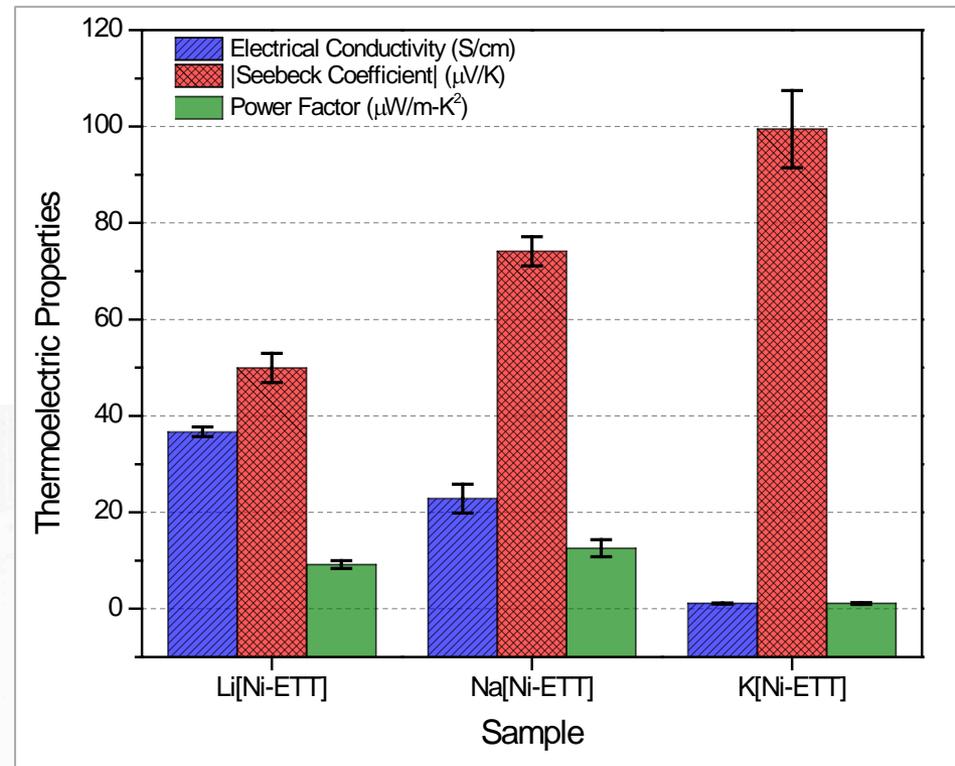


Annealing leads to impurity removal and film densification.

Synthesis Modifications (Step 1): Varying the Counterion



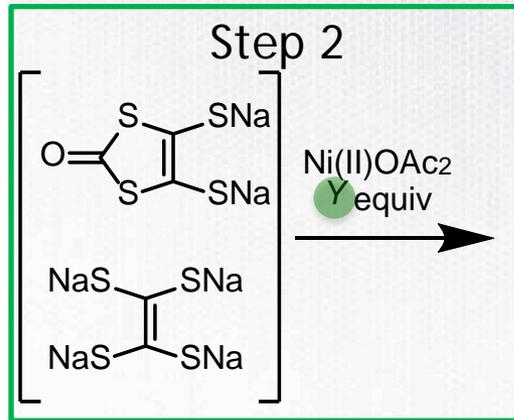
Size of the counterion
changes TE properties:
Na(NiETT) shows the
highest performance



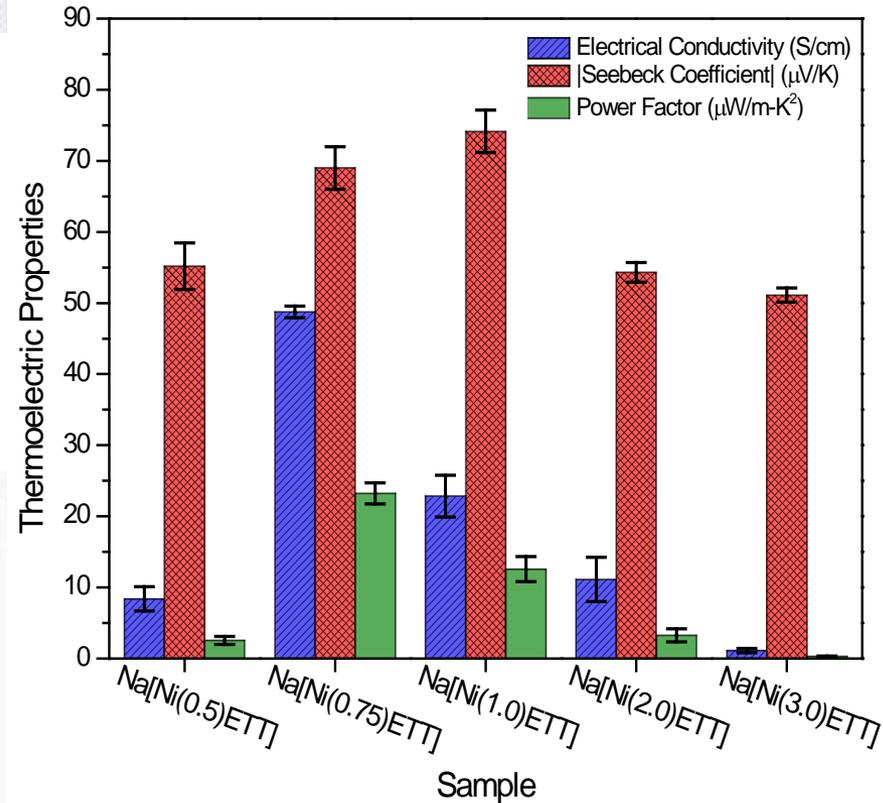
Smaller metal counter-ion = Larger conductivity

Larger metal counter-ion = Larger Seebeck

Synthesis Modifications (Step 2): Optimizing Ni(II) Amount

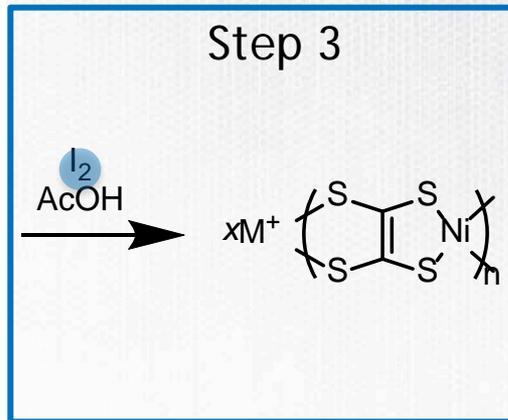


Sub-stoichiometric amounts of Ni(II) result in enhanced performance: 0.75 Ni(II) yields highest properties

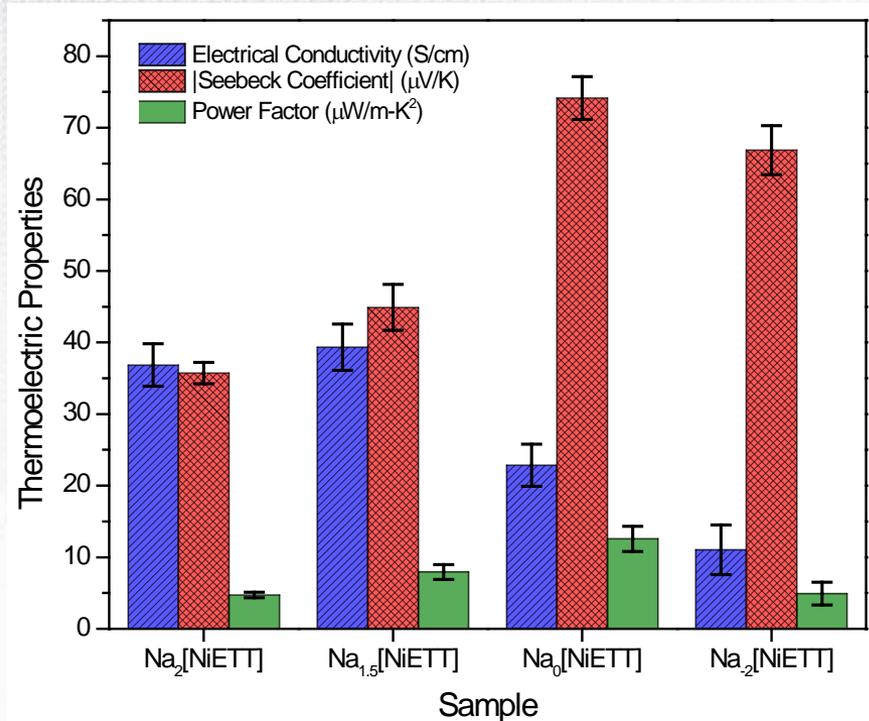


XPS reveals that the presence of excess Ni results in the formation of secondary products such as NiSO_4 and Ni(OH)_2 .

Synthesis Modifications (Step 3): Tuning the Oxidation Level

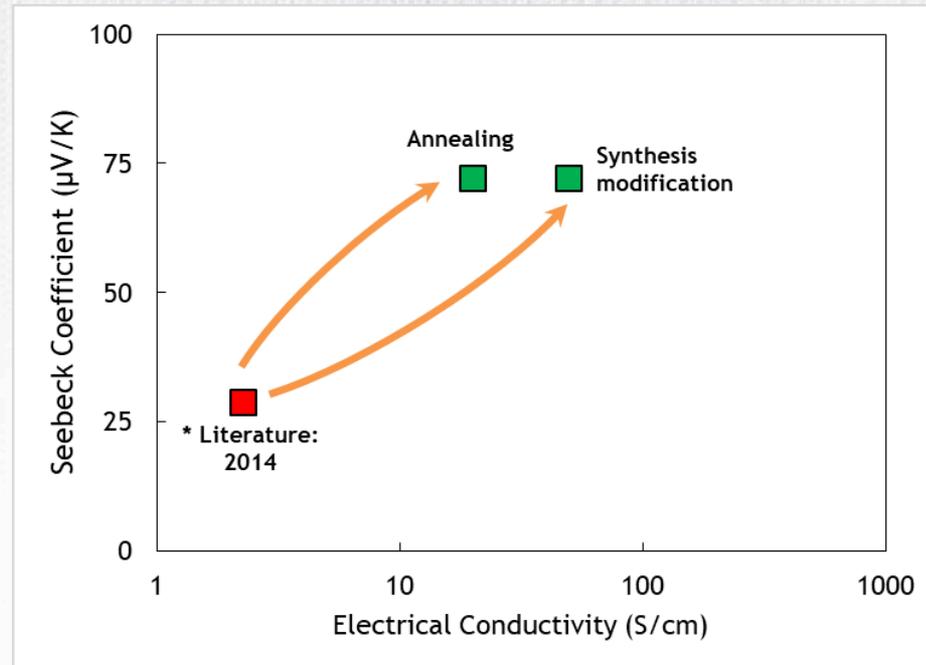


Iodine can tune the oxidation level by extracting electrons from the polymer backbone



XPS reveals that the oxidation is a ligand-centered process.

Development of a High Performance and Air Stable n-type Polymer

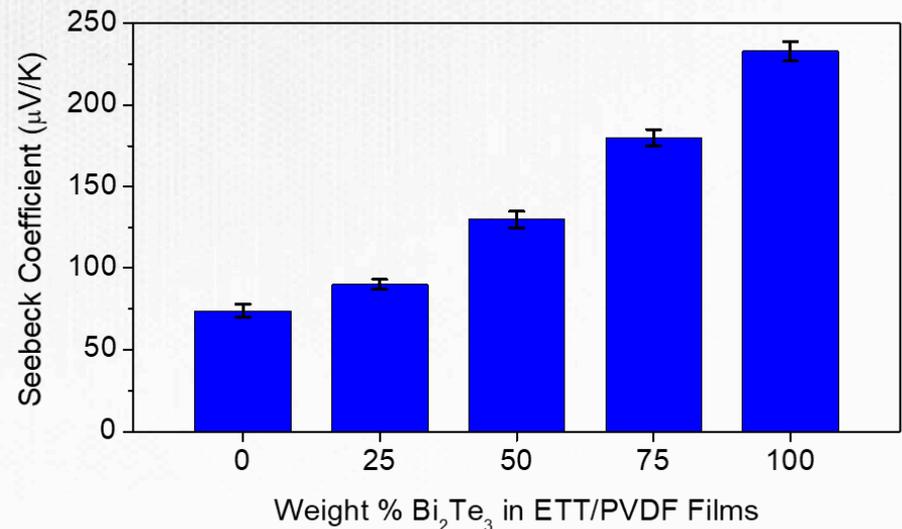
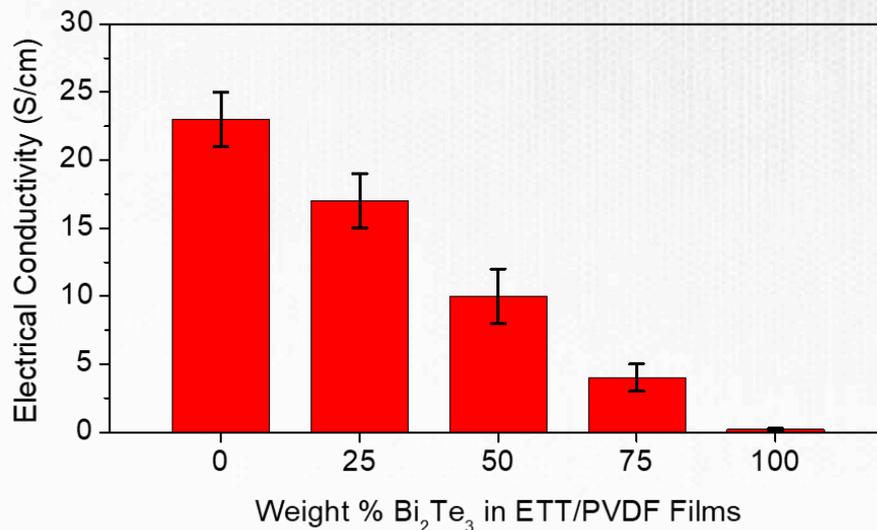


* F. Jiao et. al., *Phil Trans R Soc A*, 372, 2014.

Highest σ for a solution processed n-type film, maintains air stability, tunable viscosity for printing applications.

Enabling Organic - Inorganic Composites

Development of an air stable n-type polymer with high conductivity enables composites with a high σ (from NiETT) and high S (from Bi_2Te_3)



50 wt.% n-type Bi_2Te_3 in ETT/PVDF films results in

$$\sigma = 10 \text{ S/cm and } S = -130 \mu\text{V/K.}$$

This enables printable organic-inorganic composites.

Part IV

THERMOELECTRIC DEVICES

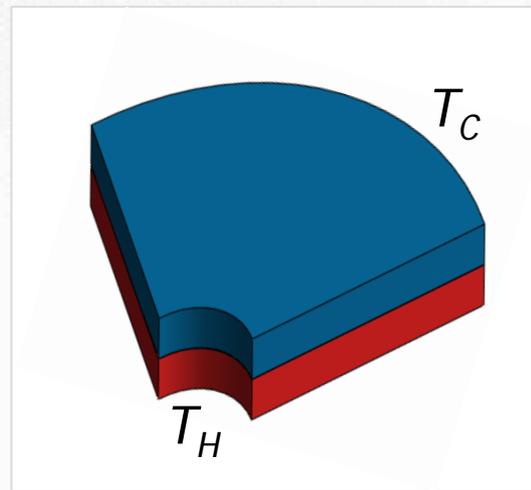
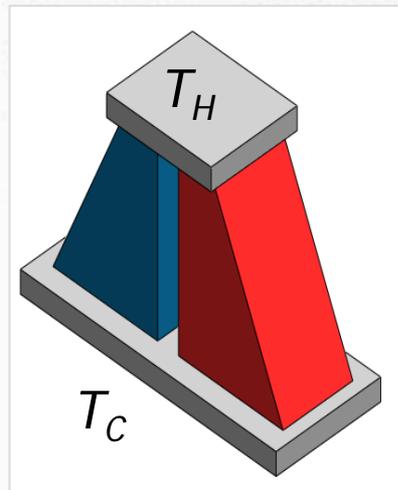
New Architectures for Polymer TEGs

$$\$/W \longrightarrow F > 1$$

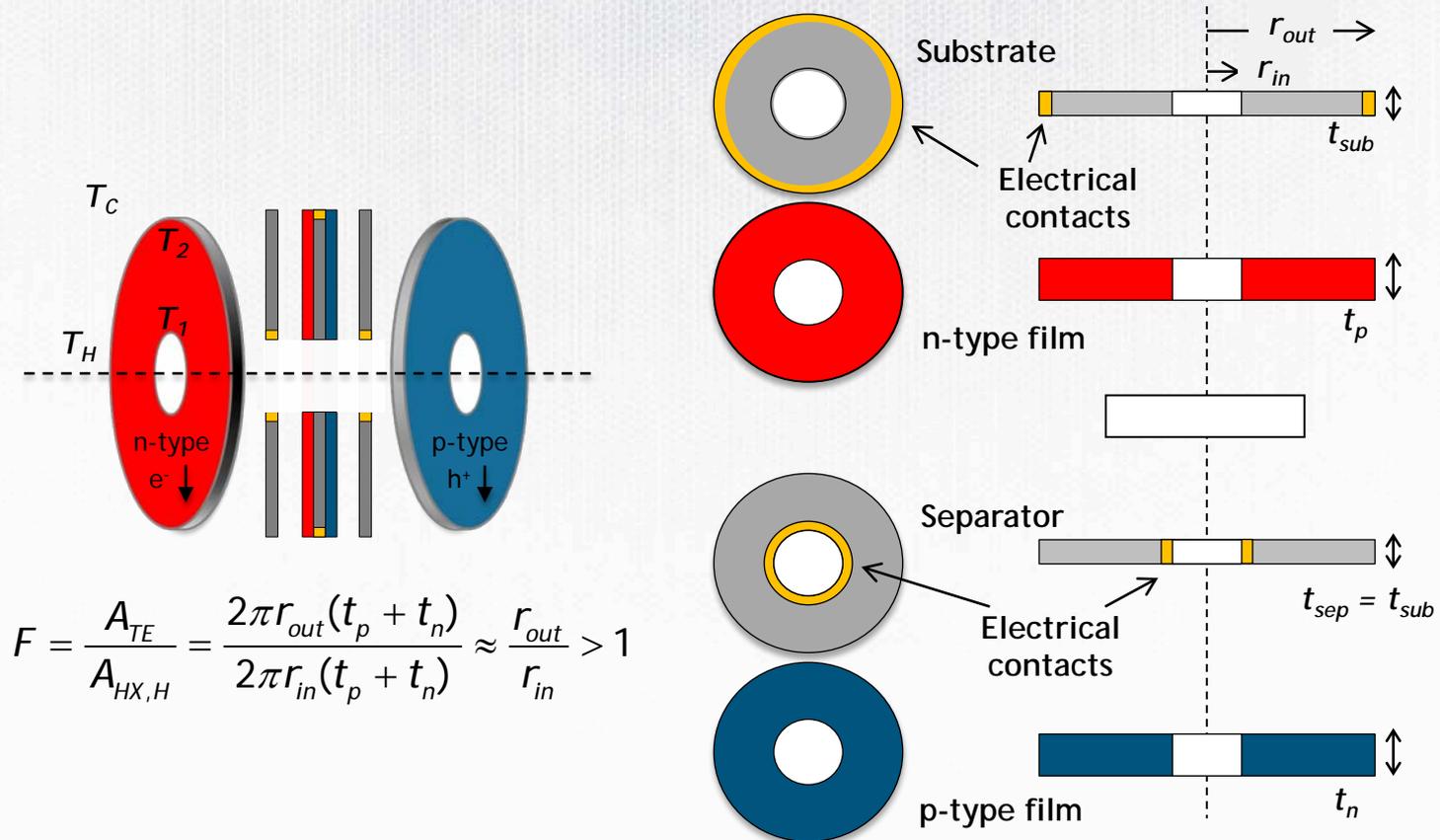
Yee et. al., *Energy Environ. Sci.*, 6, 2013.

Cooled by natural convection alone

Heat spreading



Radial TEG Concept



Conditions for Maximum Efficiency and Module ZT

- Relates device geometry with material properties
- Maximizes ZT and device efficiency

Module ZT

$$ZT = \frac{S_{pn}^2 T}{R_{el} K_{th}}$$

	Flat-plate	Radial
Electrical Resistance, R_{TE}	$\frac{L_p}{\sigma_p A_p} + \frac{L_n}{\sigma_n A_n}$	$\frac{\ln(r_{out,p} / r_{in,p})}{2\pi \sigma_p t_p} + \frac{\ln(r_{out,n} / r_{in,n})}{2\pi \sigma_n t_n}$
Thermal Conductance, K_{TE}	$\frac{k_p A_p}{L_p} + \frac{k_n A_n}{L_n}$	$\frac{2\pi k_p t_p}{\ln(r_{out,p} / r_{in,p})} + \frac{2\pi k_n t_n}{\ln(r_{out,n} / r_{in,n})}$
Geometry Matching Condition	$\frac{A_n l_p}{A_p l_n} = \sqrt{\frac{k_p \sigma_p}{k_n \sigma_n}}$	$\frac{t_n \ln(r_{out,p} / r_{in,p})}{t_p \ln(r_{out,n} / r_{in,n})} = \sqrt{\frac{k_p \sigma_p}{k_n \sigma_n}}$

Optimum Leg Length for Maximum Power Density

Includes heat exchangers to obtain temperature distribution across device

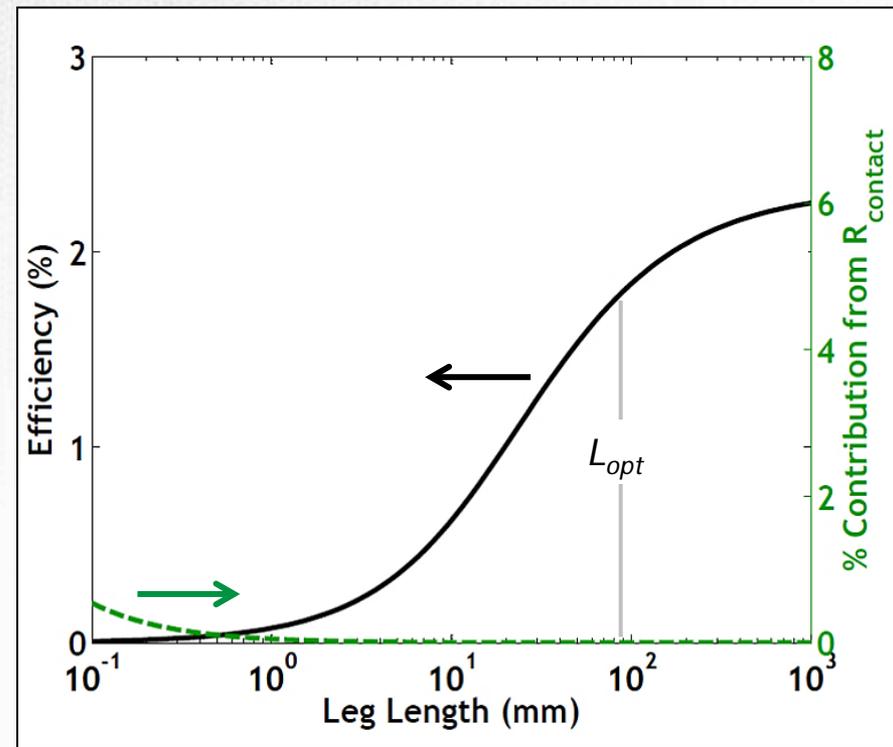
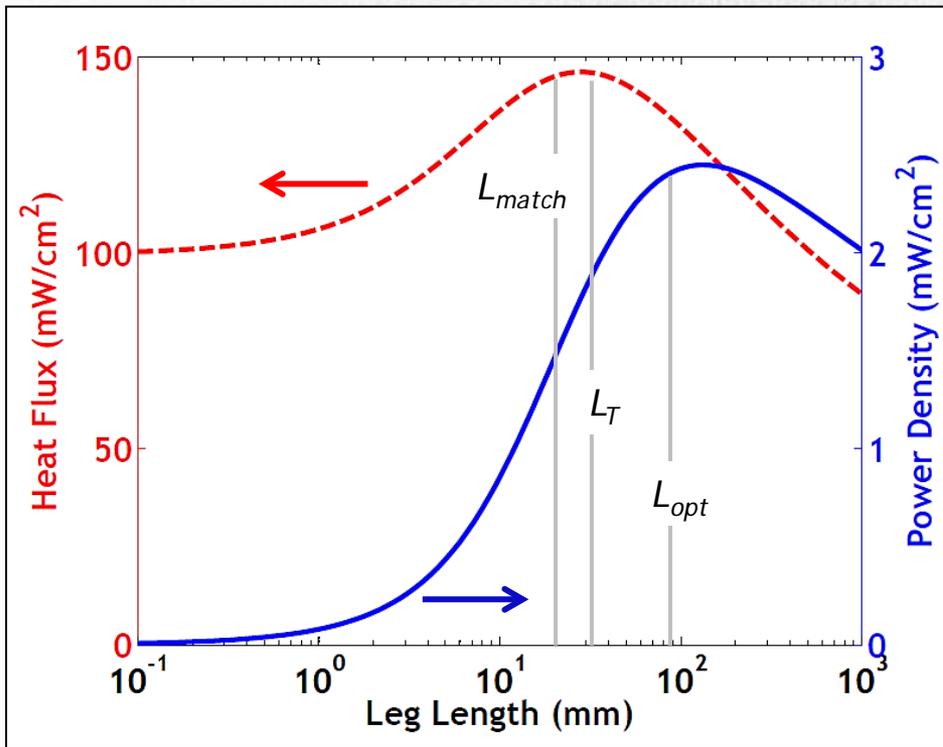
	Flat-plate	Radial
Thermal Impedance Matching Condition L_{match}	$L = 2FL_T$	$\ln\left(\frac{r_{out}}{r_{in}}\right) = \left(\frac{L_T}{r_{out}}\right)$
Optimum Leg Length L_{opt}	$L \approx 2FL_T$	$\ln\left(\frac{r_{out}}{r_{in}}\right) \approx \left(\frac{L_T}{r_{out} - 2L_T}\right)$

For radial TEG, $L_{opt} > L_{match}$ making it advantageous to operate under natural convection

Radial TEG is Advantageous for Thin-Film Devices

*Dedoped DMSO-mixed PEDOT:PSS

$S = 72 \mu\text{V/K}$, $\sigma = 890 \text{ S/cm}$, and $k = 0.33$



Negligible electrical contact resistance due to larger area of contact

Radial TEG Proof-of-Concept

- p-type¹ - PEDOT:PSS with Te nanowires
- n-type^{2,3} - Poly[K_x(Ni-ett)] blended with PVDF/DMSO

<i>p-type zT values at 300 K</i>	
Polymer Thin-Film ¹	0.1
Paper composite	0.15 x 10 ⁻³

<i>n-type zT values at 300 K</i>	
Polymer Thin-Film ³	0.2
Paper composite	0.3 x 10 ⁻³



Substrates: copy paper
Separators: polyimide



Paint p- and n-type polymers



Paint electrodes
(silver ink)

¹ Yee et. al., *Phys. Chem. Chem. Phys.*, 15, 2013.

² Sun et. al., *Adv. Mater.*, 24, 2012.

³ Jiao et. al., *Phil. Trans. R. Soc. A*, 372, 2014.

Experimental Setup

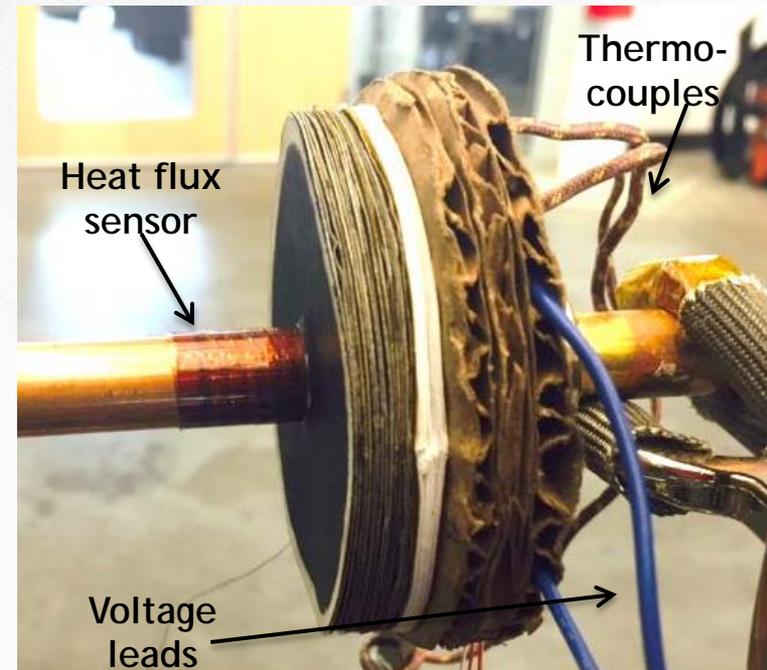
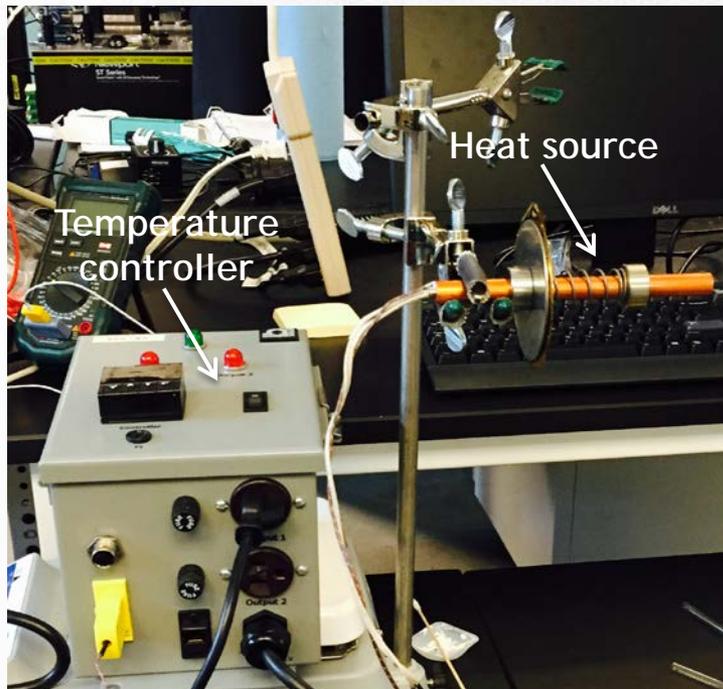


$$r_{out} = 35 \text{ mm}$$

$$r_{in} = 6.5 \text{ mm}$$

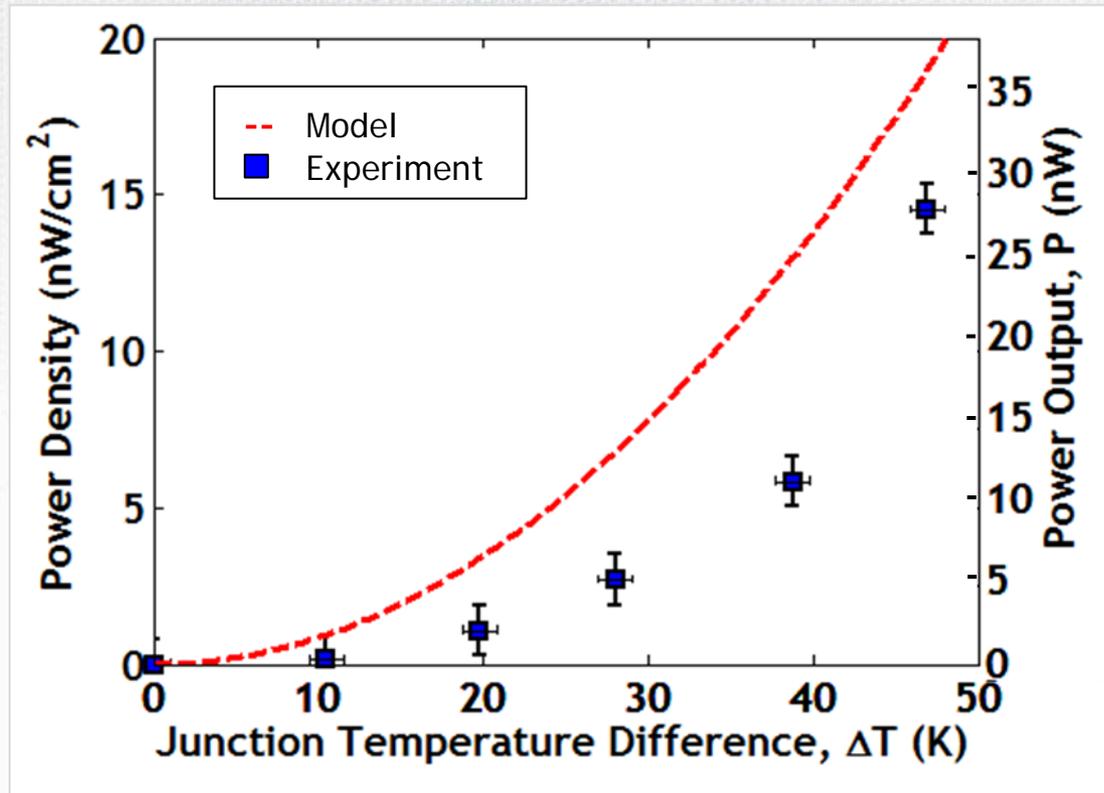
$$t_{sub} = 100 \text{ } \mu\text{m}$$

$$t_{sep} = 25 \text{ } \mu\text{m}$$



Device Performance

15 p-n junctions



Max. power output = 27 nW and power density = 14.5 nW/cm²

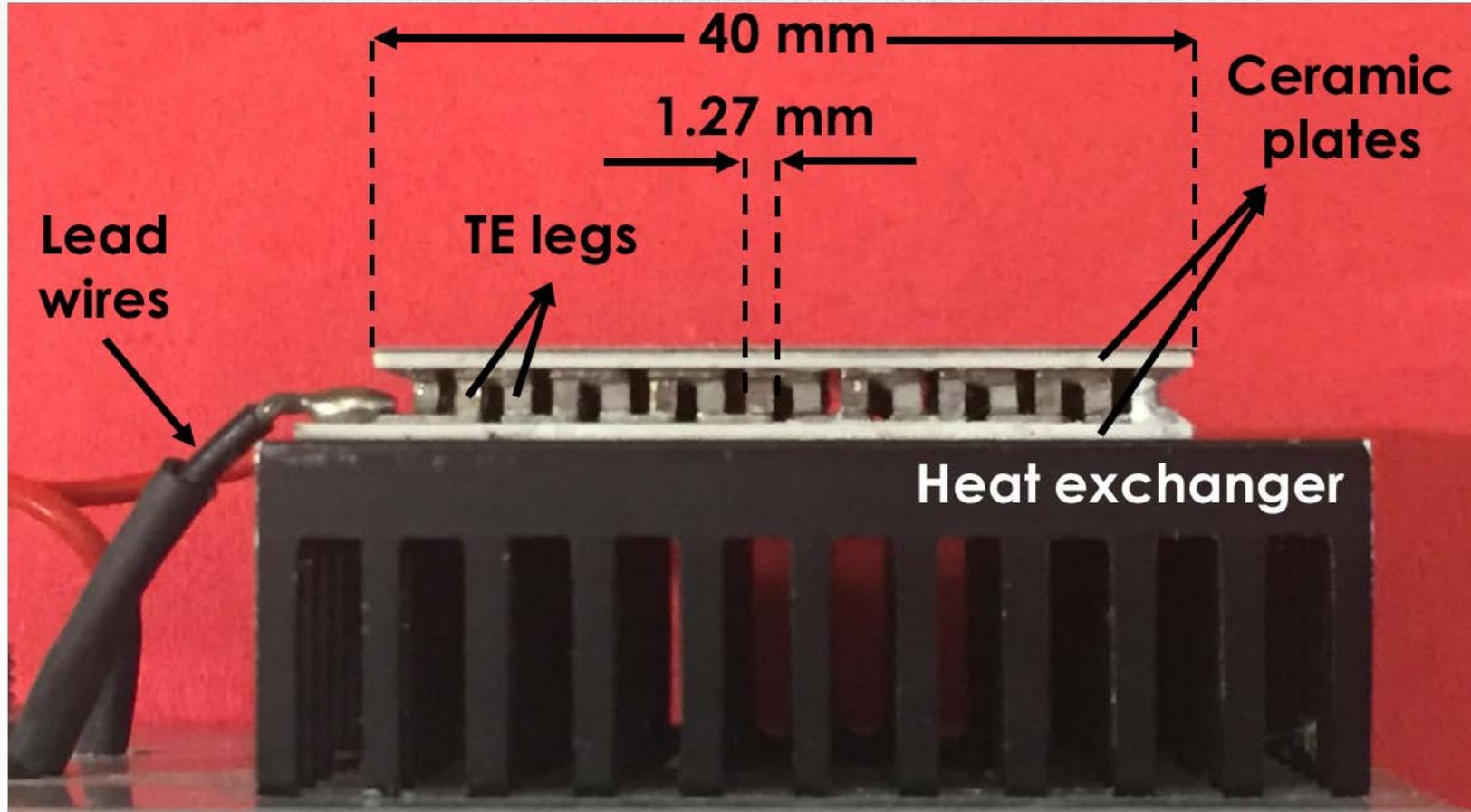
Max. open circuit voltage = 85 mV/K

First demonstration of a radial polymer TEG

Part V

PRINTED THERMOELECTRICS & TEXTILE-INTEGRATED THERMOELECTRICS

Conventional Inorganic Thermoelectric Module



FF=0.25

Leg Length ~1.5 mm

Comparing Inorganic and Polymer Thermoelectrics

Inorganics - Bi_2Te_3

$$\sigma \approx 1000 \text{ S/cm}$$

$$S \approx 200 \text{ } \mu\text{V/K}$$

$$k \approx 1.2 \text{ W/m-K}$$

$$S^2\sigma \approx 4000 \text{ } \mu\text{W/m-K}^2$$

$$zT \approx 0.8$$

Costs

Material: \$125/kg

Manufacturing: \$165/m²



2-3x more couples

HX considerations

$$\eta \leq \frac{ZT}{4} \frac{\Delta T}{T_H}$$

Low Cost
Manufacturing

Polymers - PEDOT:PSS

$$\sigma \approx 900 \text{ S/cm}$$

$$S \approx 75 \text{ } \mu\text{V/K}$$

$$k \approx 0.25 \text{ W/m-K}$$

$$S^2\sigma \approx 500 \text{ } \mu\text{W/m-K}^2$$

$$zT \approx 0.4$$

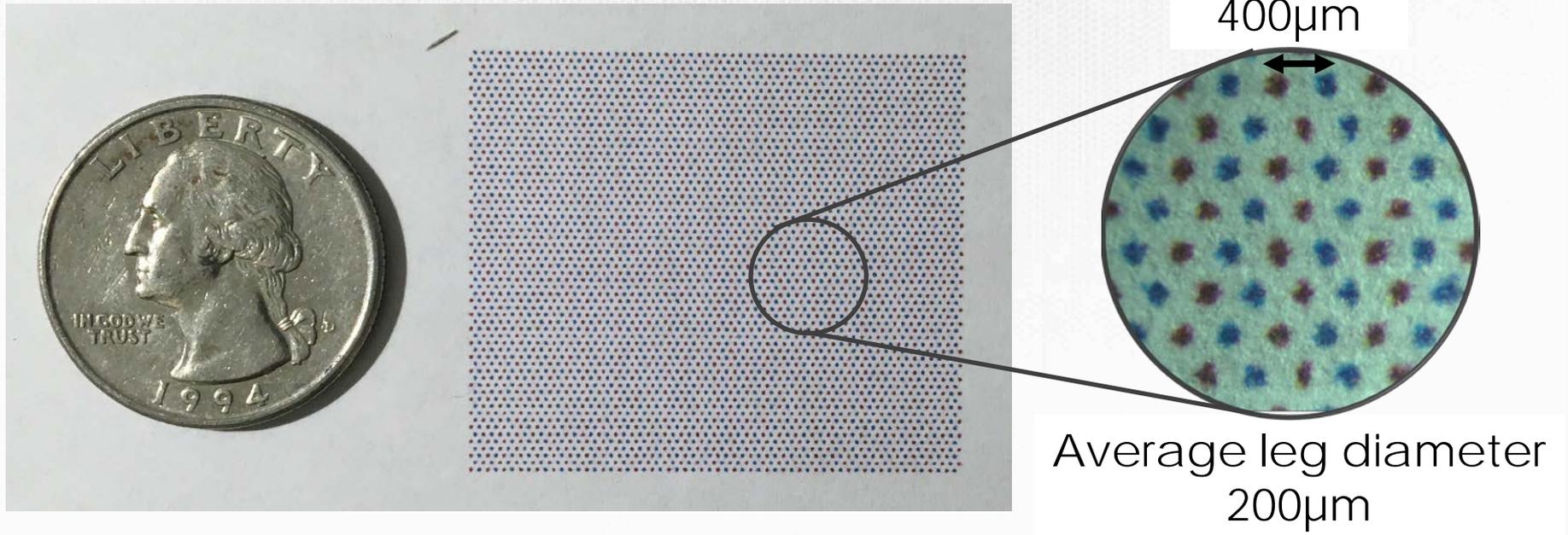
Costs

Material: \$0.34/kg

Manufacturing: \$4.76/m²



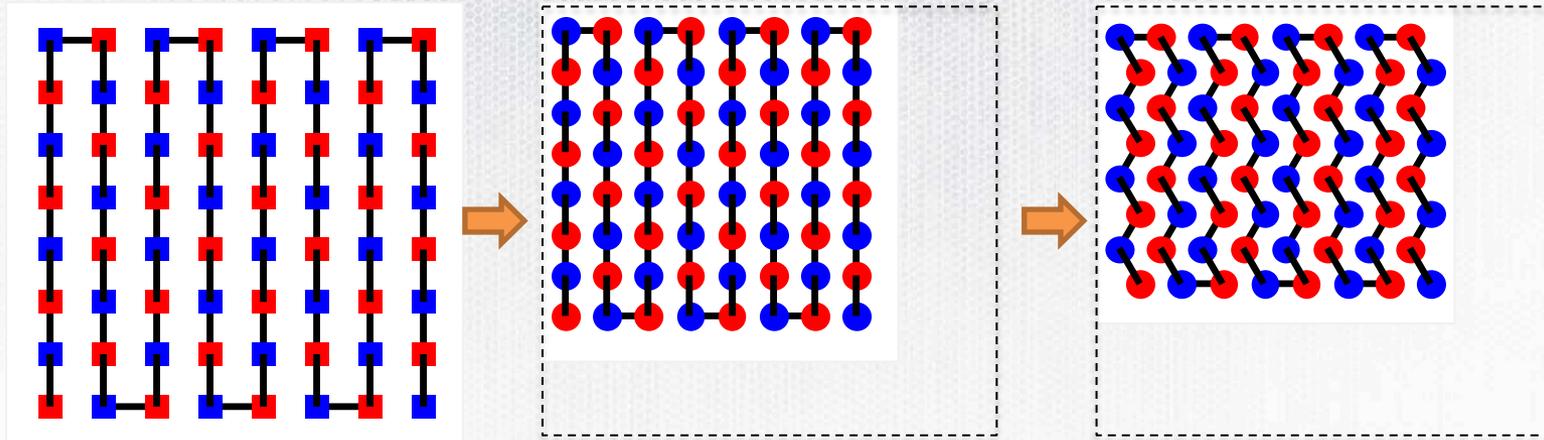
Printed Thermoelectrics



FF=0.90

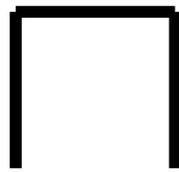
Leg Length 0.160 mm

Interconnect Patterns

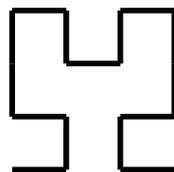


Hilbert
Curve
Order, n

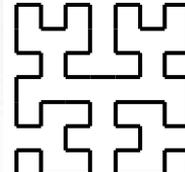
$n=1$



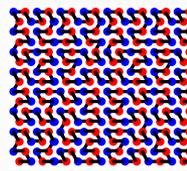
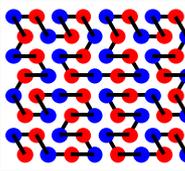
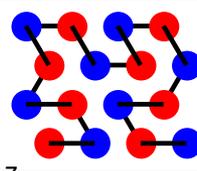
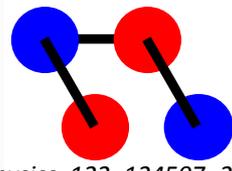
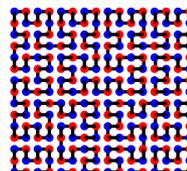
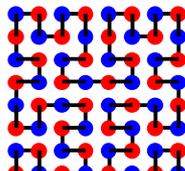
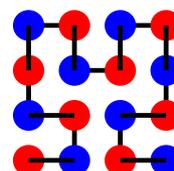
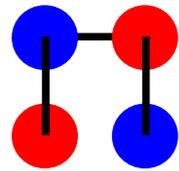
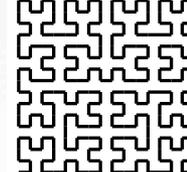
$n=2$



$n=3$

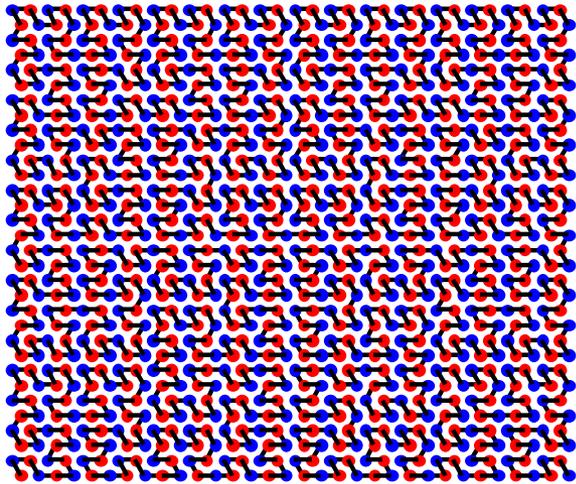


$n=4$

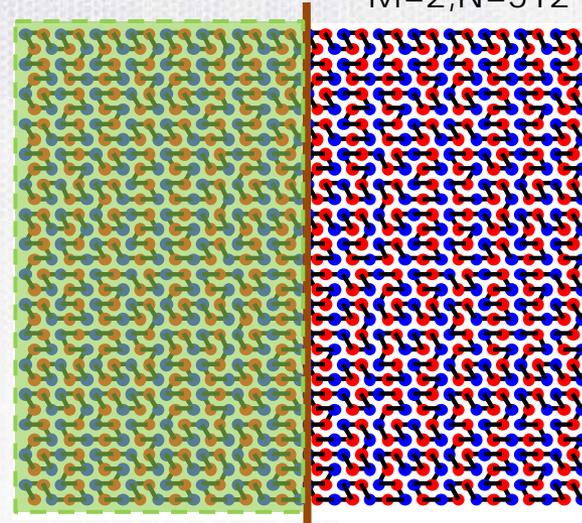


Sub-Module Localization

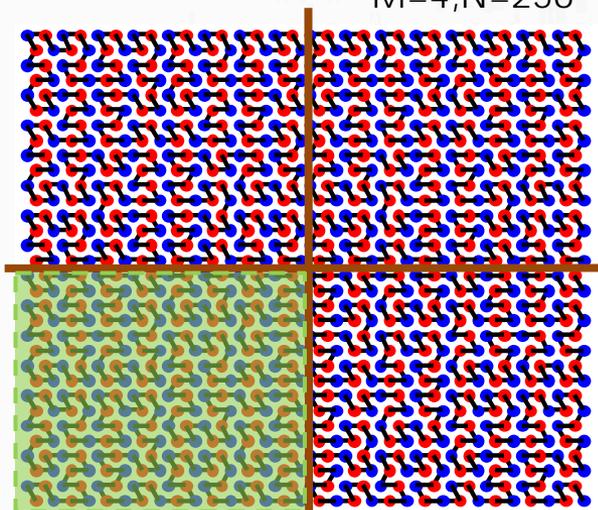
$M=1, N=1024$



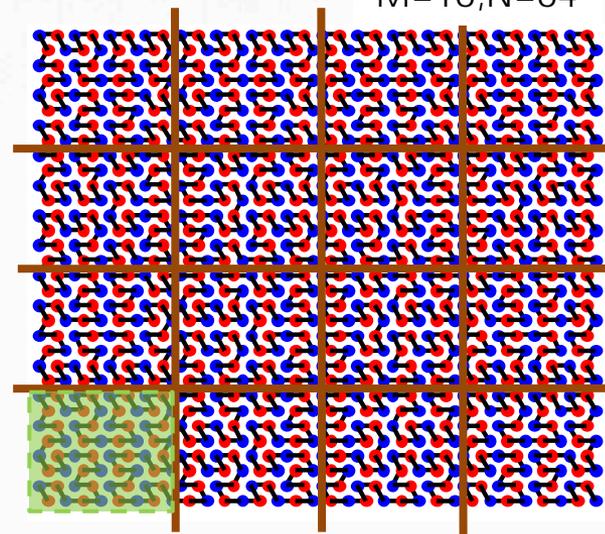
$M=2, N=512$



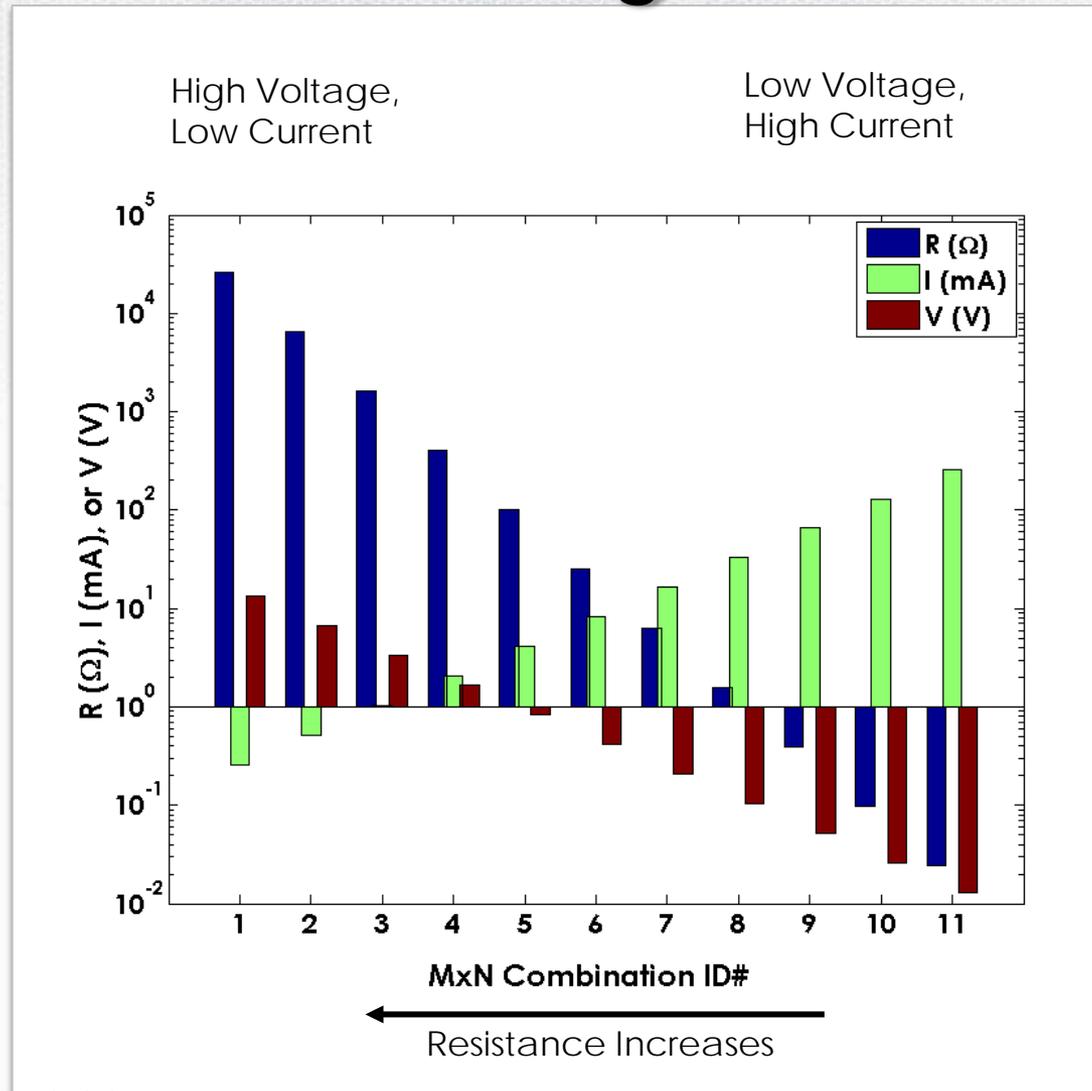
$M=4, N=256$



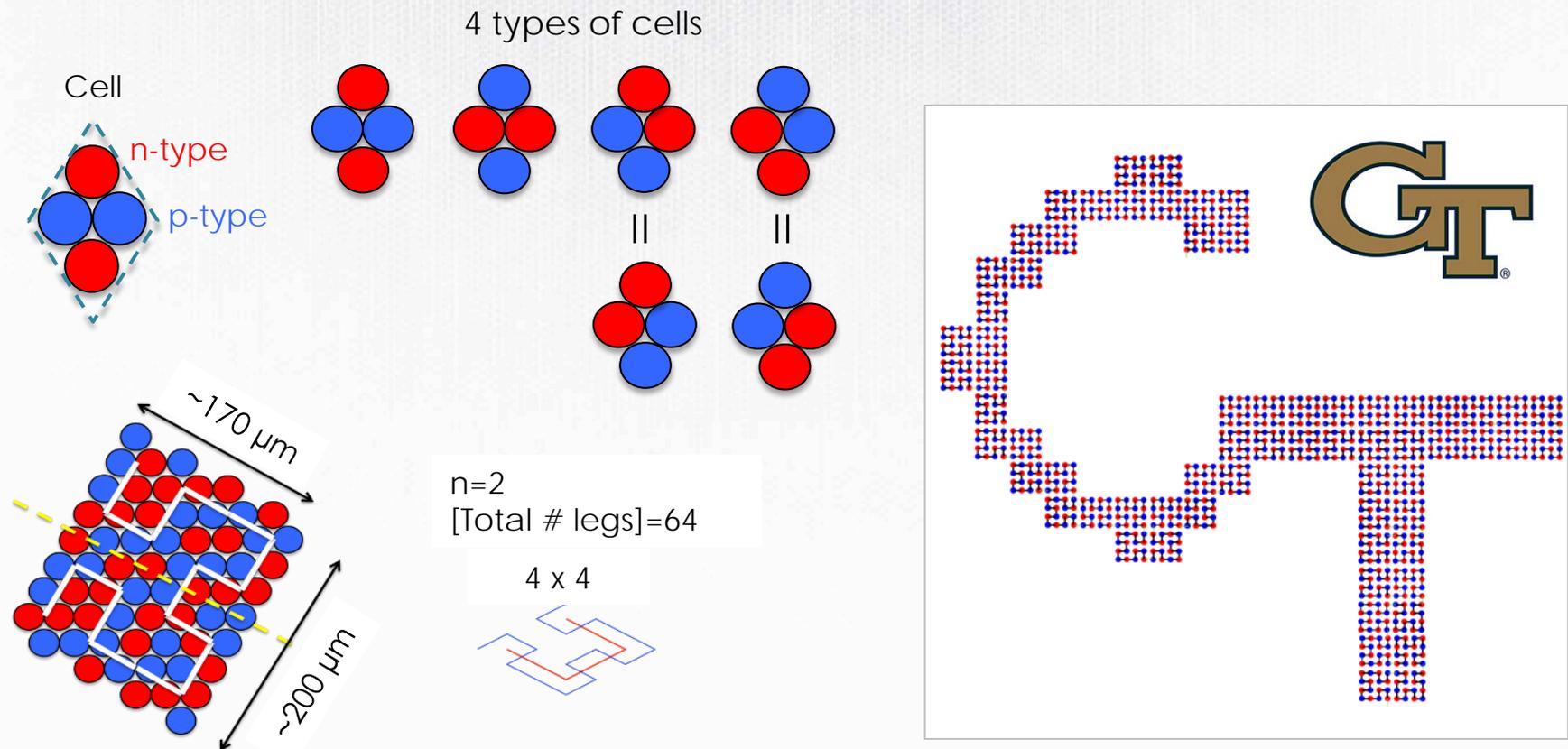
$M=16, N=64$



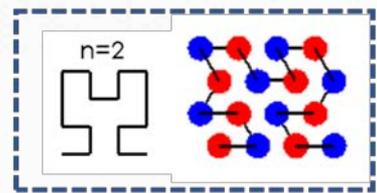
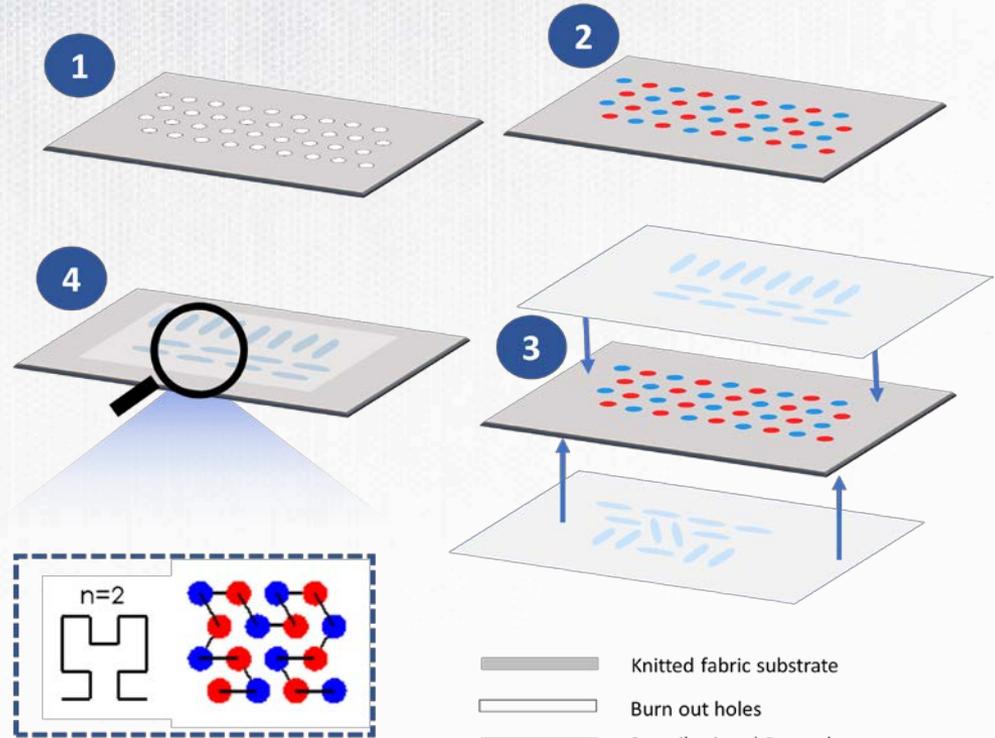
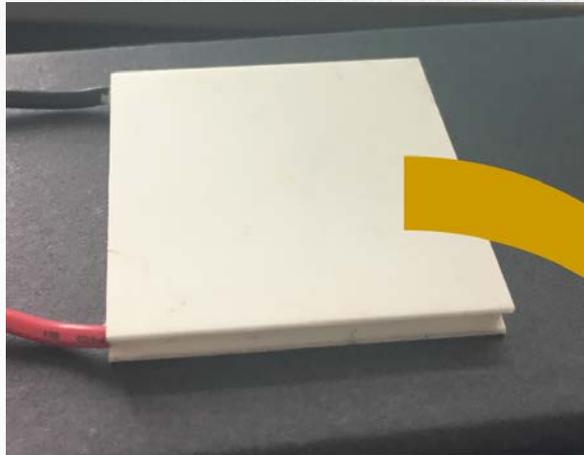
Impedance tuning avoids power conditioning circuits



Printing into any shape requires a change of basis from two-legs to four-legs

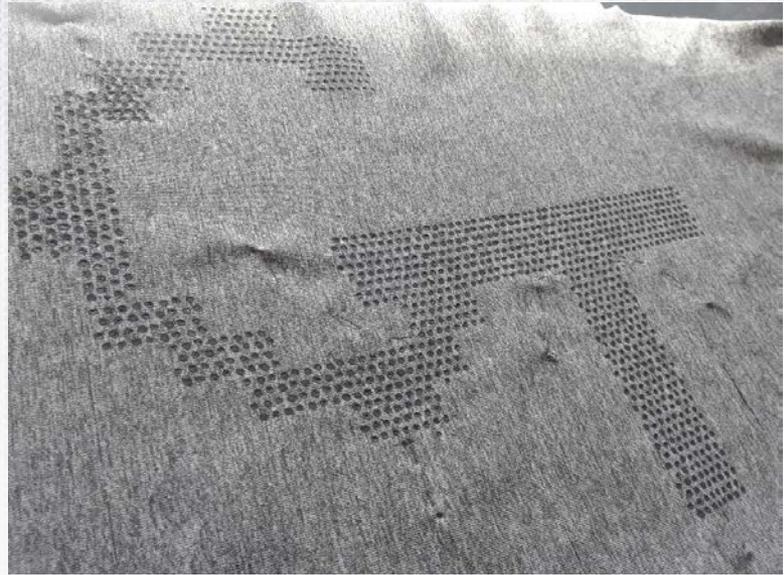


Textile-Integrated Devices



TE legs arranged in hexagonal closed-packed layout and connected by the Hilbert curve thus increasing the Fill Factor (FF=0.36)

Design Concepts: Textile-Integrated TE



To Be Continued...



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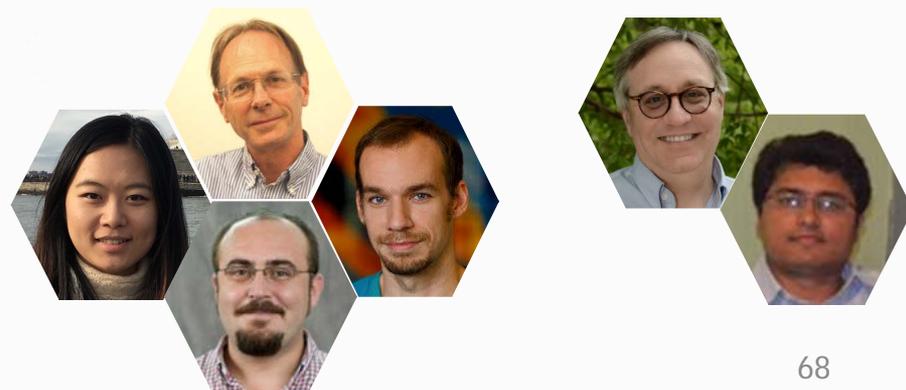
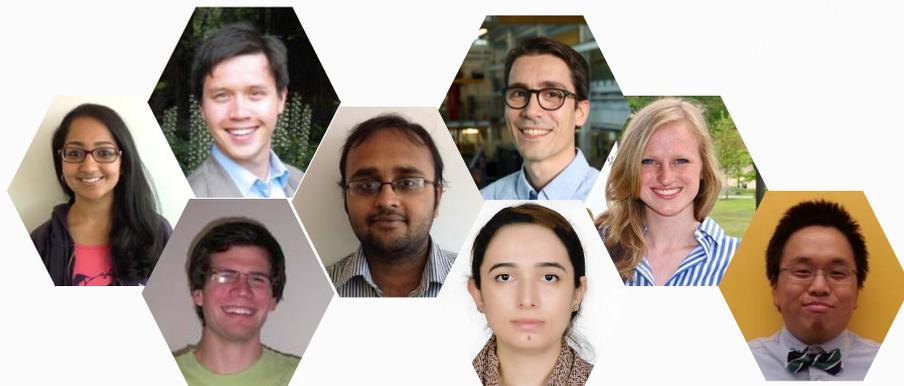
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