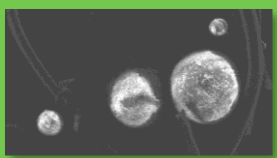
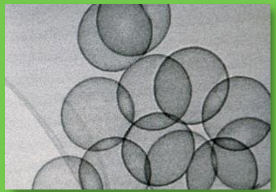
U.S. DEPARTMENT OF ENERGY
Office of ENERGY EFFICIENCY & RENEWABLE ENERGY

OAK RIDGE National Laboratory

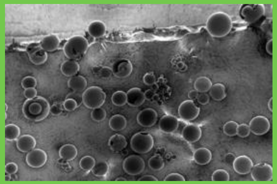
Models to Evaluate and Guide the Development of Low Thermal Conductivity Materials for Building Envelopes



ORNL's polymeric vacuum insulation spheres



ORNL's hollow silica particles



ORNL's coated and evacuated insulation spheres

New models that combine nanoscale and macroscale heat transfer will provide means to develop low thermal conductivity materials

ORNL's ultralow thermal conductivity materials

LBNL's robust super insulation

Oak Ridge National Laboratory
Som Shrestha, R&D Staff
865-241-8772, shresthass@ornl.gov

Project Summary

Timeline

Start date: 10/1/2018

Planned end date: 9/30/2021

Key Milestones

1. Collect input from advisory board, 3/31/19
2. Develop and refine models, 9/30/2019
3. Calibrate models using measured data, 9/30/2020
4. Design options to achieve ≥R14/in., 9/30/2021

Key Partner:

Vanderbilt University

Budget

Total Project $ to Date:

- DOE: $240K
- Cost share: 0

Total Project $:

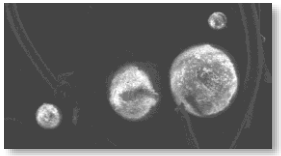
- DOE: $850K
- Cost share: 0

Project Outcome

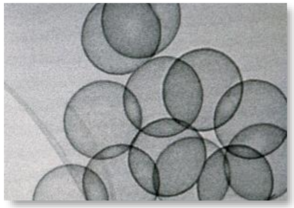
Models that combine nanoscale and macroscale heat transfer will guide researchers in the development of thermal insulation materials with R-values ≥14/in.

Challenge

- BTO has a keen interest in insulation materials with ≥R14/in. (thermal conductivity (TC) ≤0.01 W/mK)
- Researchers are attempting to reduce effective thermal conductivity by applying heat transfer phenomena that have shown to decrease thermal conductivity in semiconductors, such as interfacial thermal resistance (ITR) and subcontinuum thermal transport (STT)
- Data on ITR and STT and models that incorporate these phenomena are not available for most amorphous materials with properties that are favorable to achieve high R/inch



ORNL's polymeric vacuum insulation spheres



ORNL's hollow silica particles

ORNL's ultralow thermal conductivity materials

LBNL's robust super insulation

R/in.: thermal resistivity, h.ft^2.°F/Btu/inch

Objective

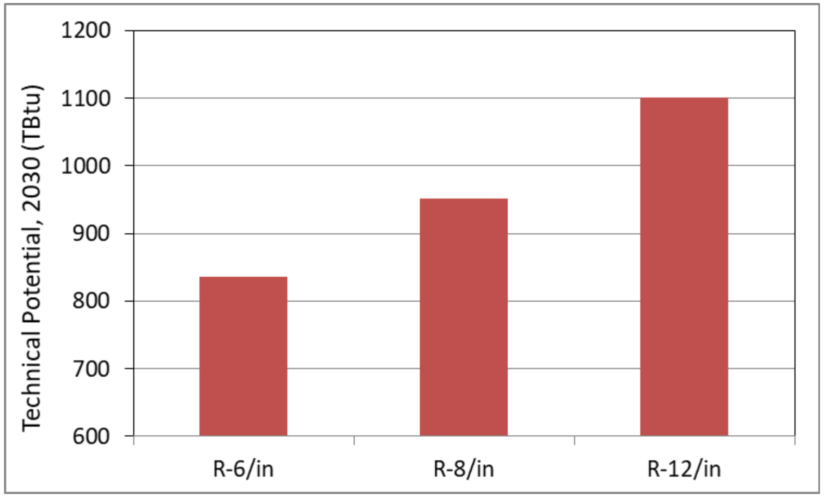
Develop models that capture interfacial thermal resistance, subcontinuum thermal transport, and other relevant heat transfer phenomena, and translate the nanoscale/microscale effects to the macroscale level that is relevant to building insulation materials

- Integrate advances in semiconductor research
- Apply latest developments in molecular dynamics (MD) simulations

Significant Potential Impact

- Available models for insulation materials do not capture heat transfer phenomena that have been studied for semiconductors and that require molecular dynamics simulations

- The proposed modeling tool will capture all modes of heat transfer through complex structures, which will expand methods to develop new insulation materials



Technical potential for 2 in. of insulation material[1]

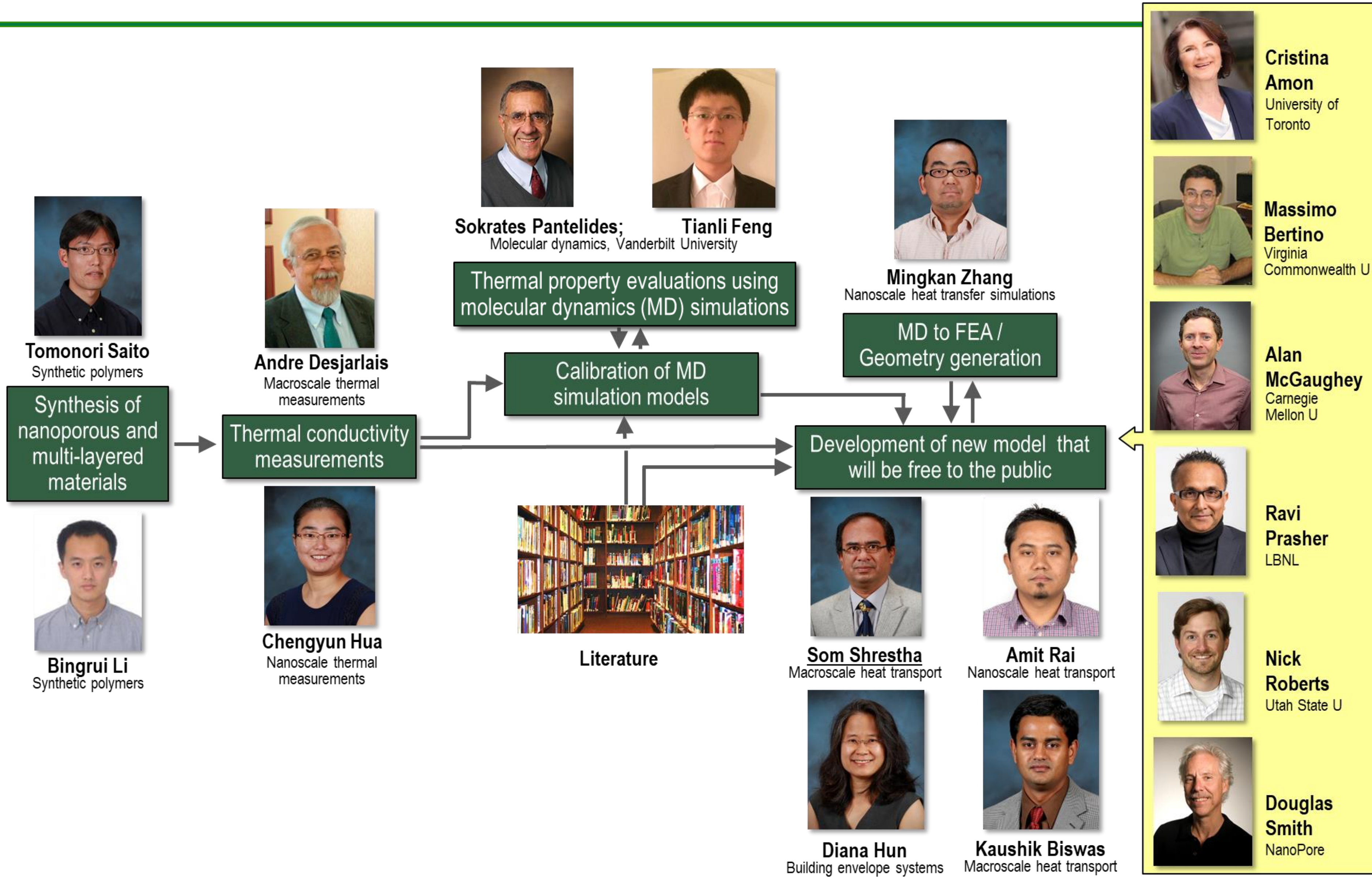
- Technical potential for R12/in. insulation materials is 1.1 Quads[1]

Customers

✓ Direct: researchers developing new insulation materials
✓ Indirect: insulation materials manufacturers

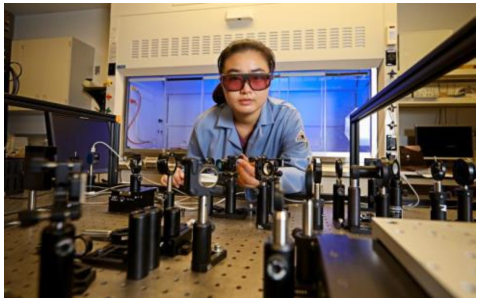
[1] DOE BTO Windows and Building Envelope R&D: Roadmap for Emerging Technologies, Feb 2014. In 2030, assuming addition of 2-in. insulation.

Team

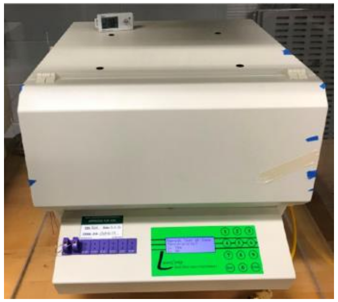

Advisory Board
Cristina Amon
University of Toronto
Massimo Bertino
Virginia Commonwealth U
Alan McGaughey
Carnegie Mellon U
Ravi Prasher
LBNL
Nick Roberts
Utah State U
Douglas Smith
NanoPore
Sokrates Pantelides; Tianli Feng
Molecular dynamics, Vanderbilt University
Thermal property evaluations using molecular dynamics (MD) simulations
Mingkan Zhang
Nanoscale heat transfer simulations
MD to FEA / Geometry generation
Tomonori Saito
Synthetic polymers
Synthesis of nanoporous and multi-layered materials
Bingrui Li
Synthetic polymers
Andre Desjarlais
Macroscale thermal measurements
Thermal conductivity measurements
Chengyun Hua
Nanoscale thermal measurements
Calibration of MD simulation models
Literature
Development of new model that will be free to the public
Som Shrestha
Macroscale heat transport
Amit Rai
Nanoscale heat transport
Diana Hun
Building envelope systems
Kaushik Biswas
Macroscale heat transport

Resources at ORNL

- Material scientists and Center for Nanophase Materials Sciences expertise for synthetizing samples needed to measure ITR and STT
- Expertise in heat transfer modeling and measurements
- High-performance computing to perform large numbers of complex MD simulations



Time-domain thermoreflectance (TDTR)



Heat flow meter apparatus (HFMA)

Transient plane source (TPS)

HFMA with controlled pressure and gas composition

ITR: interfacial thermal resistance   STT: subcontinuum thermal transport

Advisory Board

Objectives

- Get feedback on our modeling approach
- Ensure completeness of our plan
- Improve usefulness and credibility of our models

Cristina Amon	University of Toronto	Computational nanoscale thermal transport
Alan McGaughey	Carnegie Mellon University	
Nick Roberts	Utah State University	
Massimo Bertino	Virginia Commonwealth University	• Nanoscale thermal transport • Development of high R/in. materials
Ravi Prasher	LBNL	
Douglas Smith	NanoPore	

- First meeting on Mar 20, 2019
- Next meeting in mid-June, 2019

Overall Approach

Gather and integrate feedback from advisory board

FY19 Model development	FY20 Model calibration	FY21 Optimization
Develop and integrate physics-based and empirical models • Bulk heat transfer • STT • ITR • Morphology	Evaluate key variables that can be controlled in the lab • Pore shape, size, and wall thickness • Gas conductivity • Gas pressure • Temperature • Interface density	Tune key variables that could significantly affect thermal conductivity • Base material • Binders • Pore shape and size • Pore wall thickness • Gas pressure • ITR

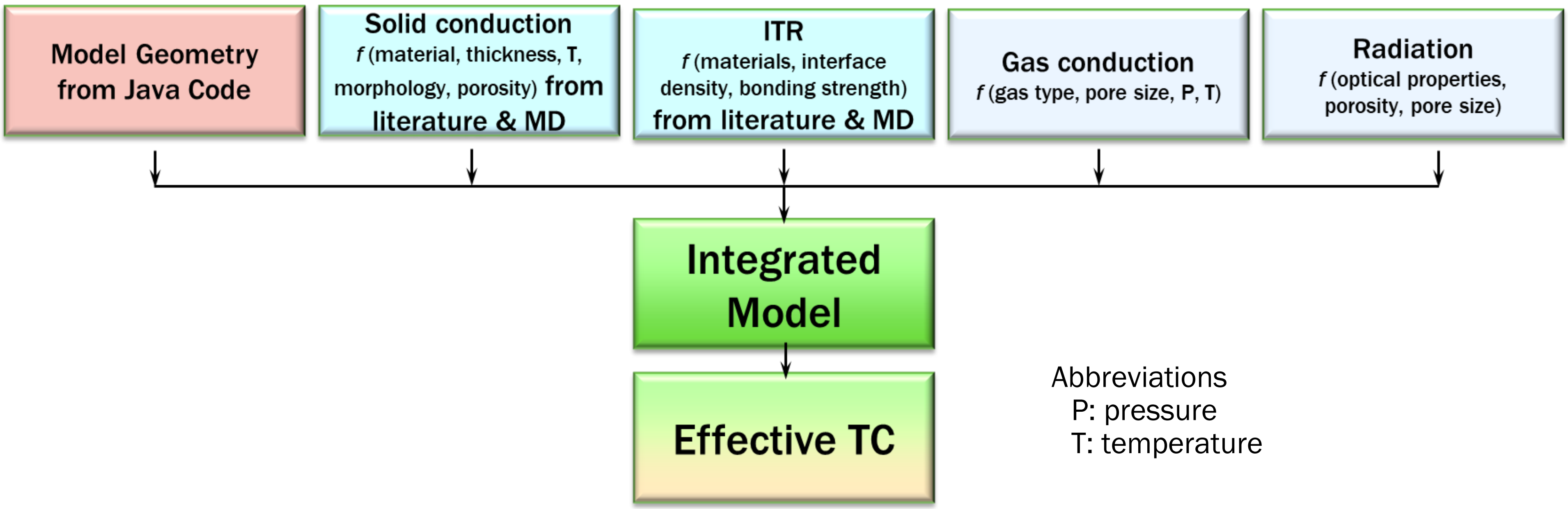
ITR: interfacial thermal resistance

STT: subcontinuum thermal transport

Approach: Model Development

Develop integrated physics-based and empirical models that capture

- Macroscale and nanoscale heat transfer
- Interfacial thermal resistance
- Morphology



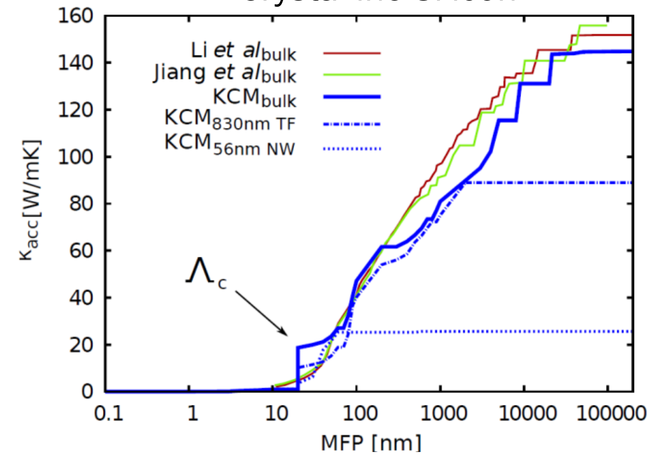
Abbreviations
P: pressure
T: temperature

Approach: Expand Existing Research

Subcontinuum thermal transport

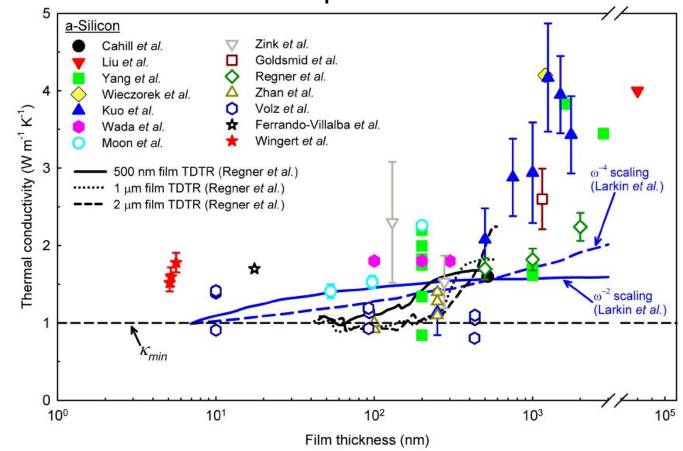
- Available research focuses mainly on crystalline materials
  - Effective TC decreases with size reduction
- Limited research on amorphous materials
  - Lower TC than crystalline materials
  - Impact of STT on amorphous polymers is not well studied
- Assess impact of STT on TC of amorphous polymers that have inherently lower TC with MD simulations and experiments

Crystalline Silicon



Accumulated TC as a function of mean free path with different size, Alvarez (2018)

Amorphous Silicon



Wingert et al. (2016)

Significant reduction in TC as the thickness reduces ***but still high to achieve desired low TC***

Approach: Expand Existing Research (cont.)

Interfacial thermal resistance

- Available research is primarily on metals or polymers + crystalline materials
- Limited research on amorphous polymers with lower TC → better suited to achieve high R/inches
- Conduct MD simulations and experiments to identify types of interfaces with high thermal resistance

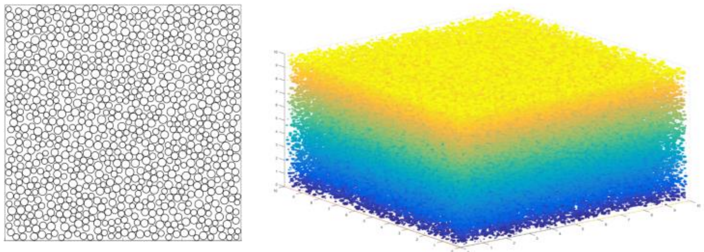
Sample ITR data	ITR		Source
	$m^2.K/W$	$h.ft^2.°F/Btu$	
Polymer thin film and aluminum thin film	1.0E-07	5.7E-07	J. Liu, (2013)
Silica/germanium	2.5E-09	1.4E-08	Feng et al. (2019)
Rhodium/magnesium oxide	1.0E-08	5.7E-08	Prasher and Phelan (2001)

Requires millions of interfaces per inch thickness to get useful R-values for building insulation purposes

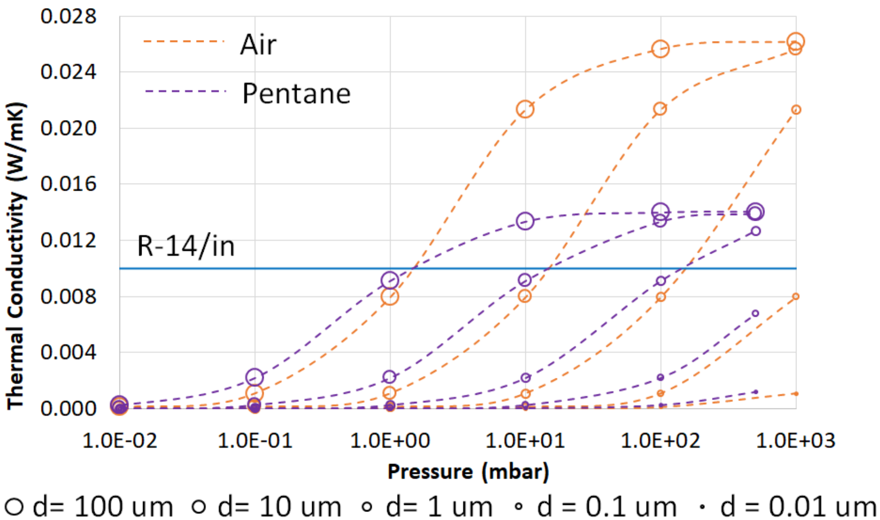
Progress: Model Development

- Generated geometry of porous materials with size and thickness distribution → potential method to integrate ITR and STT in bulk materials consisting of layered or stacked nanomaterials
- Integrating gas conduction as a function of pore size, porosity, and pressure

Coloring is just to make easier to view individual spheres



Randomly distributed 2D and 3D objects with different sizes, porosity, and wall thickness



Gas TC as a function of pore size and pressure using kinetic theory of gases

Progress: Molecular Dynamics Simulations

Aim: Predict the effect of

- Solid-solid interfacial resistance $R_{s\text{-}s}\ (\varepsilon, T)$
- Impact of STT on solid TC: in- and through-plane $K_{s,xyz}\ (L, T)$

for materials that could potentially be used to achieve high R/inch

Progress

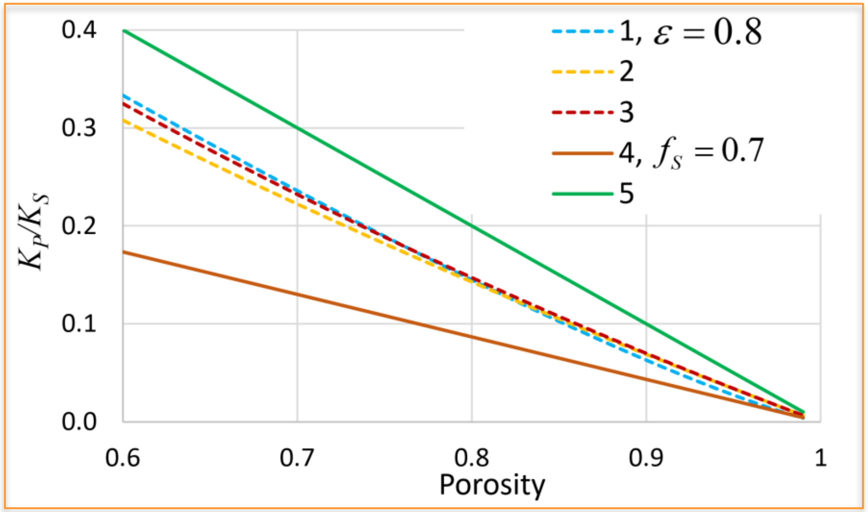
- Started MD simulations for polystyrene and benchmarked its bulk TC

Amorphous PS	Atoms	λ, W/mK
Our MD simulation	3,366	0.156
	26,928	0.159
Experimental data		0.14 ~ 0.19

ε – bonding strength
L – characteristic length (thickness)
T – temperature
λ - thermal conductivity

Progress: Impact on Solid Conduction due to Porosity

- Various methods used to evaluate the impact on solid conduction due to porosity
- Reviewing the validity of these models when the thickness of the materials is comparable to the mean free path of the energy carrier



1. Effective medium approximation

$$\frac{K_P}{K_S} = (1-p)^{\frac{3\varepsilon}{2}}$$

2. Eucken model

$$\frac{K_P}{K_S} = \frac{1-p}{1+p/2}$$

3. Russell model

$$\frac{K_P}{K_S} = \frac{1-p^{2/3}}{1-p^{2/3}+p}$$

4. For foams using fraction of solid in struts

$$\frac{K_P}{K_S} = \left(\frac{2}{3}-\frac{f_S}{3}\right)(1-p)$$

5. Simplified method for hollow spherical material

$$\frac{K_P}{K_S} = 1-p$$

K_P – contribution to the effective conductivity due to solid conduction
K_S – solid TC

p – porosity
ε – shape factor, 1 for spheres
f_S – fraction of solid in the foam struts

Progress: Options to Calculate Radiation HT

Planck's law used to calculate electromagnetic radiation might not be applicable to calculate radiative heat transfer in pores with diameters smaller than the infrared radiation wavelength (~8 to 25 μm at room temperature)

Option 1: Simplified models that use extinction coefficient

Challenge: limited extinction coefficient data.

Compiling what is available in the literature

Option 2: Detailed models using near-field thermal radiation for pore sizes smaller than infrared wavelength

Options suggested by the Advisory Board

Remaining Project Work: FY 19–20

- Calculate size and morphology-dependent TC and ITR for up to four amorphous polymers that have inherently lower TC using MD simulations
- Integrate size and morphology-dependent TC, ITR, gas conduction, and radiation into models to predict the effective TC of bulk materials

Remaining Project Work: FY 20–21

- Calibrate models to improve accuracy
  - Thickness-dependent thermal conductivity
  - Interfacial thermal resistance
  - R-value of porous insulation materials at various conditions
    - Air and at least two other gases
    - Various pressure levels to check the accuracy of the developed models with regard to gas conduction
    - Various temperature differentials or default vs. low-emissivity surface on heat flow meter apparatus to check the accuracy of the developed models to capture radiation heat transfer
- Use the calibrated models to generate guidelines for the development of new insulation materials with low thermal conductivity

Thank You

Oak Ridge National Laboratory

Som S. Shrestha
shresthass@ornl.gov

865-241-8772

Visit our website: www.ornl.gov/buildings

Follow us on Twitter: @ORNLbuildings

REFERENCE SLIDES

Project Budget

Project Budget: $350K in FY 19
Variances: None.
Cost to Date: $82K
Additional Funding: None

Budget History					
FY 2018 Past		FY 2019 (current)		FY 2020 – FY2021 (planned)	
DOE	Cost-share	DOE	Cost-share	DOE	Cost-share
$130K	0	$220K	0	$500K	0

Project Plan and Schedule

Project Schedule												
Project Start: 10/1/2018	Completed Work											
Projected End: 9/30/2021	Active Task (in progress work)											
	Milestone/Deliverable (Originally Planned)											
	Milestone/Deliverable (Actual)											
	FY2019				FY2020				FY2021			
Task	Q1 (Oct-Dec)	Q2 (Jan-Mar)	Q3 (Apr-Jun)	Q4 (Jul-Sep)	Q1 (Oct-Dec)	Q2 (Jan-Mar)	Q3 (Apr-Jun)	Q4 (Jul-Sep)	Q1 (Oct-Dec)	Q2 (Jan-Mar)	Q3 (Apr-Jun)	Q4 (Jul-Sep)
Past Work												
Reviewed models and data available in literature and confirmed that a model that allows to simultaneously vary all important input parameters described in the proposal is not publicly available												
Obtained input from an advisory board on the proposed modeling approach & developed new integrated models												
Current/Future Work												
Calibrate the models using measured data to gain confidence in the models												
Refine the models to minimize the discrepancies												
Evaluate the R-value at different pressures, temperatures and with various gas mixtures. Tune the models using measured data.												
Identify design options to achieve ≥R14/in.												