Enhancing LED Performance

—

A Case For Research on Encapsulant Materials With Enhanced Thermal Conductivity

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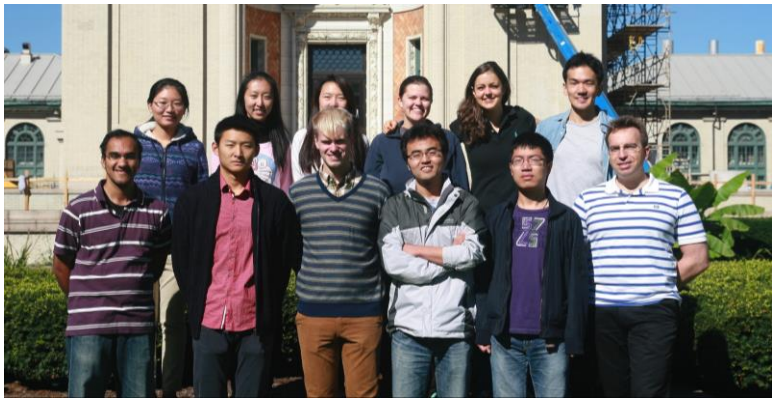
- Justin Bult (Dow Corning)



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DOE-EERE : DE-EE0006702

Polymers & LEDs

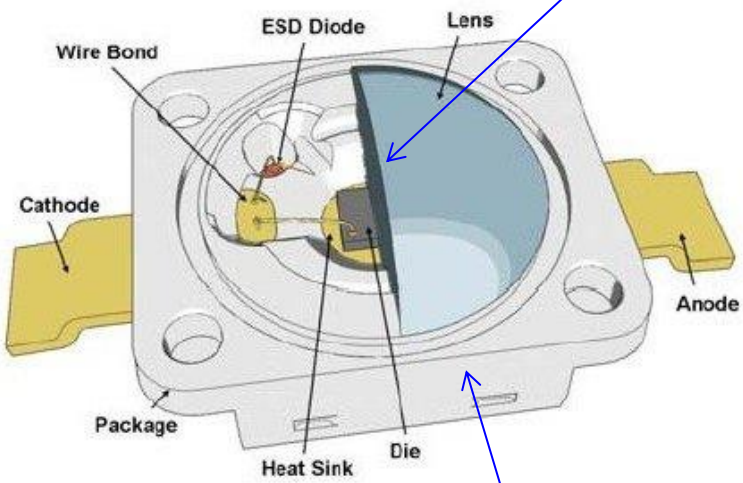
courtesy of M. Tchoul

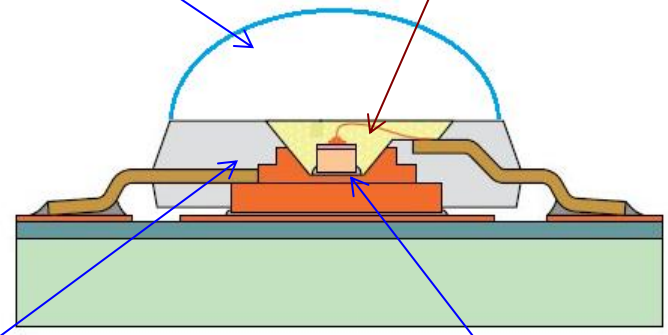
a look inside 'the package'

Package is a component in the lamp, not a lamp itself

Lens:
siloxane, epoxy, thermoplastic

Encapsulant:
siloxane, epoxy, hybrid





Housing material:
Thermoplastic, (possible siloxane)

Interconnect:
Epoxy, acrylic, solder (possible siloxane)

Transparent Siloxanes

courtesy of M. Tchoul

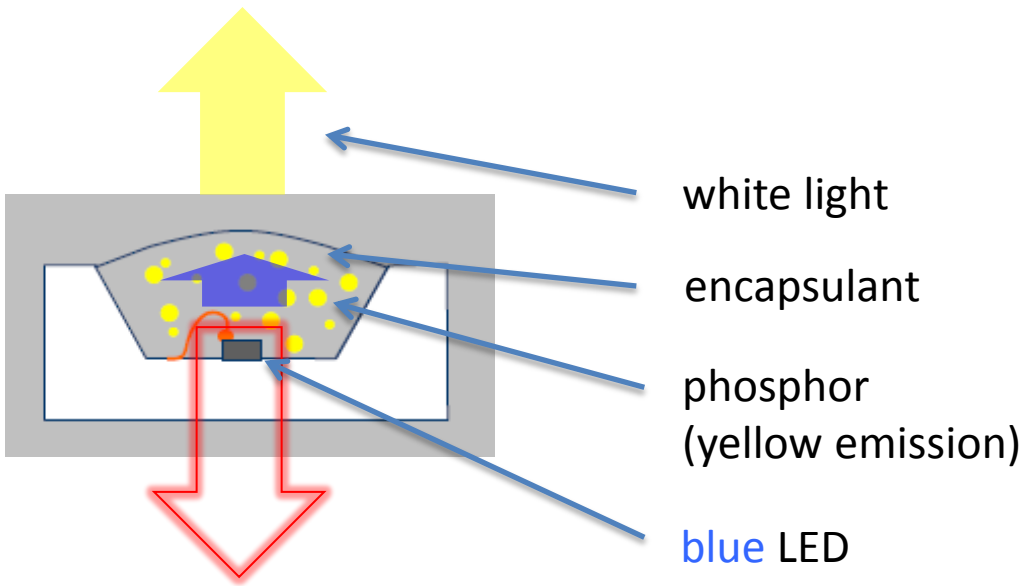
Property	Value for silicones	Desired for LED
Transmission at 1 mm	>90%	> 90 %
Temperature stability	110 - 180 *C	100-150 *C
Stability to discoloration	Moderate to high	High
Refractive index	1.46-1.54	> 1.7
Moisture permeability, g*mm/m^2*day	10-100	<1
Oxygen permeability, cc*mm/m^2*day	200-40,000	<100
Thermal conductivity	0.15-0.2 W/mk	> 1.0 W/mK
Flammability rating	HB, V-2, V-1, V-0	HB,…5-VA

Low moisture uptake ≠ low moisture permeability!

Encapsulants & pc-LEDs

source: OsramSylvania
MYPP SSL-R&D, 2014

- **phosphor-conversion LEDs represent most widely used solid-state white light system (here: volume-cast design)**



Problem

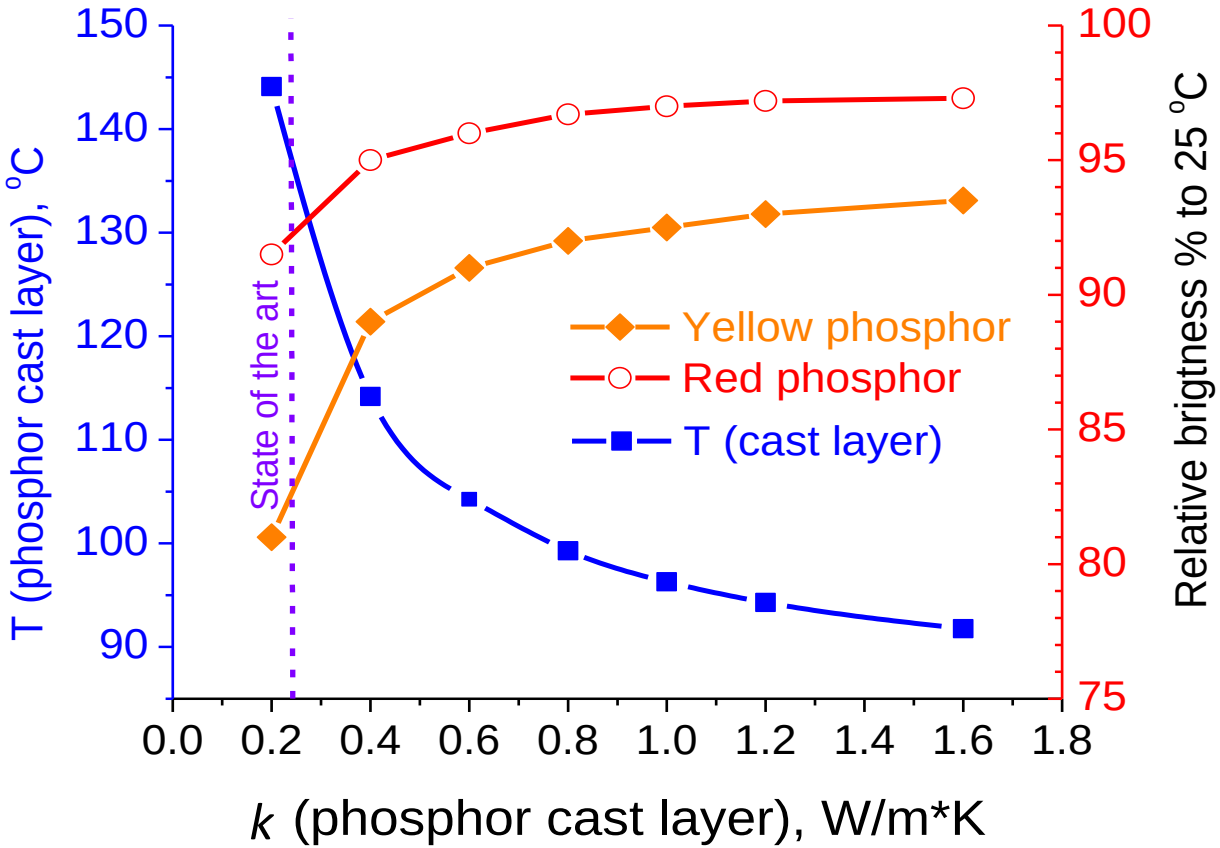
20-30% of the absorbed energy is lost because of Stokes-loss and less-than-unity QE

challenge: low thermal conductivity *k* of current siloxane encapsulants results in heating of phosphor particles and rapid degradation of conversion efficiency

Challenge of Low *k*

courtesy of OSRAM Sylvania

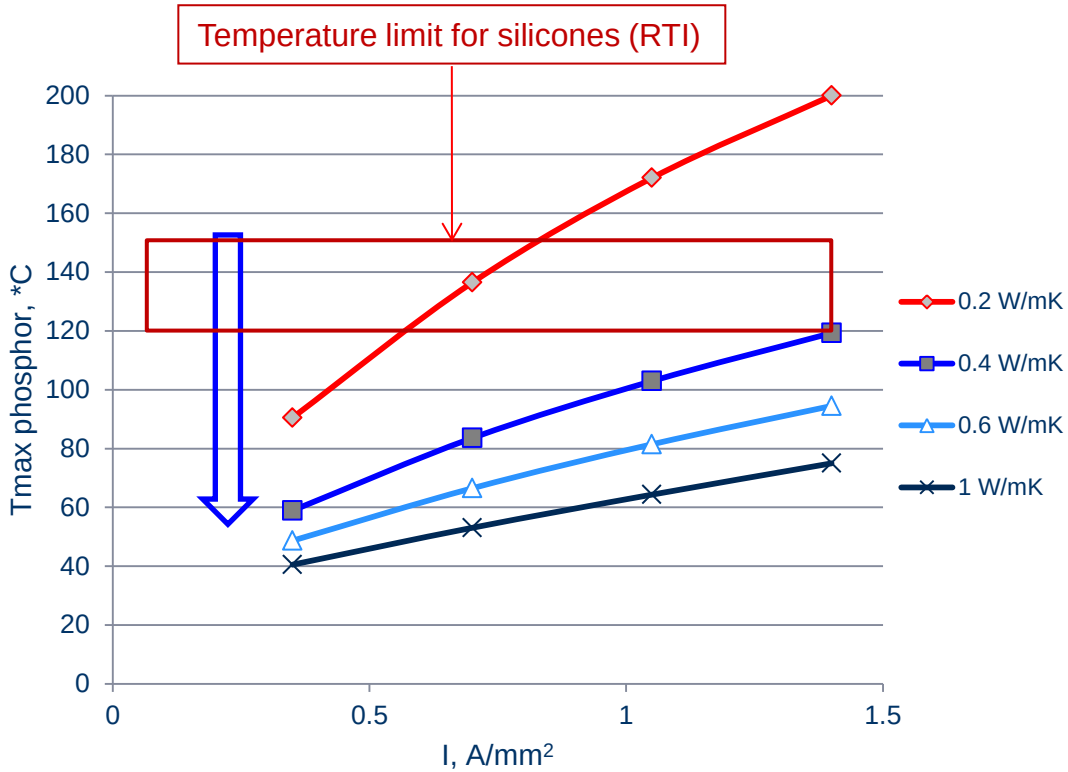
phosphors work great but …

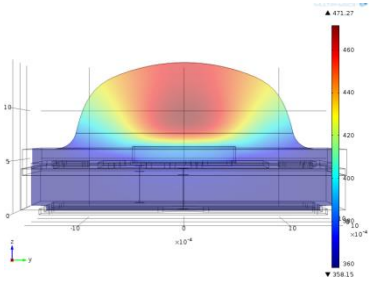


… only if "kept cool"

Opportunity

courtesy of OSRAM Sylvania





increase of *k* could enable:

higher current density, increased lifetime, lower cost, …

Background

thermal transport in polymers

critical quantity: thermal conductivity *k*

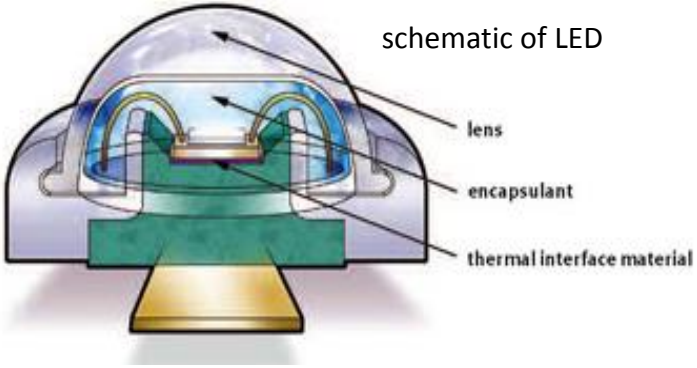
$$\dot{Q} = -k \, \mathrm{grad}(T)$$ (Fourier law)

examples

Polymers (siloxanes,…) : k ~ 0.15 W/mK
SiO_2 (amorphous) : k ~ 1 W/mK
Copper : k ~ 400 W/mK

low thermal conductivity in polymers due to phonon scattering (amorphous, free volume)

low thermal conductivity presents problem for application of polymers in packaging of high energy density 'materials or components' (energy storage, electronics, *solid state lighting*, …)



courtesy of OSRAM Sylvania

Background

strategies to raise thermal conductivity of polymers

(chain orientation)

'thermal network structures'

→ inorganic fillers

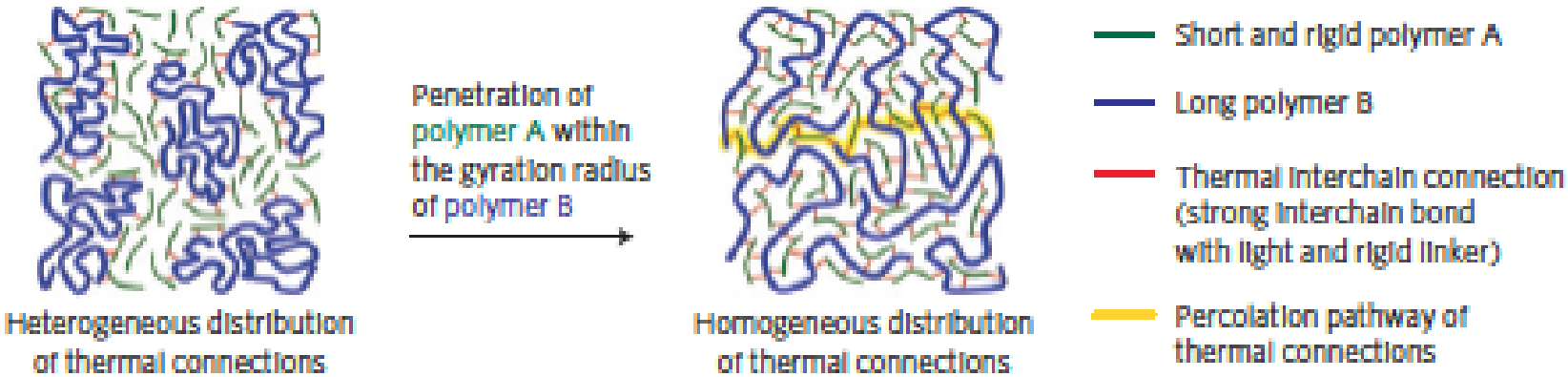
G.H. Kim *et. al. Nat. Mater.* **2015**, 14, 295-300. S. Shen *et. al. Nat. Nanotech.* **2010**, 5, 251-255.

Background

strategies to raise thermal conductivity of polymers

2. Thermal Network Structures

concept: enable secondary-bond network structures in polymer blend structures



+ thermal conductivities in excess of 1.5 W/m K demonstrated

+ optical transparency

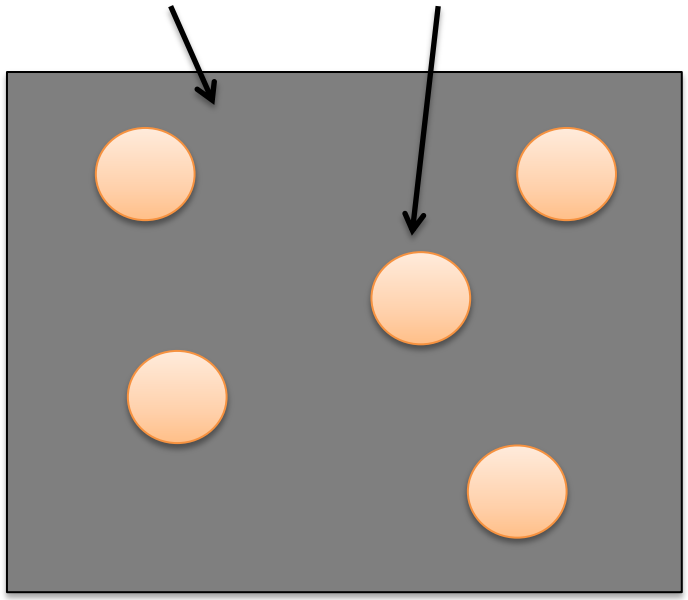
+ environmental & photothermal stability

G.H. Kim *et. al. Nat. Mater.* **2015**, 14, 295-300.

Background

raising *k* with inorganic additives

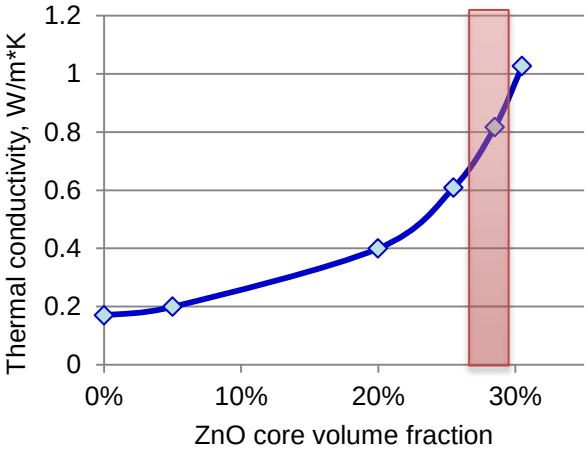
polymer matrix high *k* filler



homogenization theory

material	*k* [W/m K]
SiO_2	1.2
ZrO_2	3
Si_3N_4	20
ZnO	21
Al_2O_3	35

example:
k as function of *f*(ZnO)



if only it would be this simple !!!

source: kyocera.com

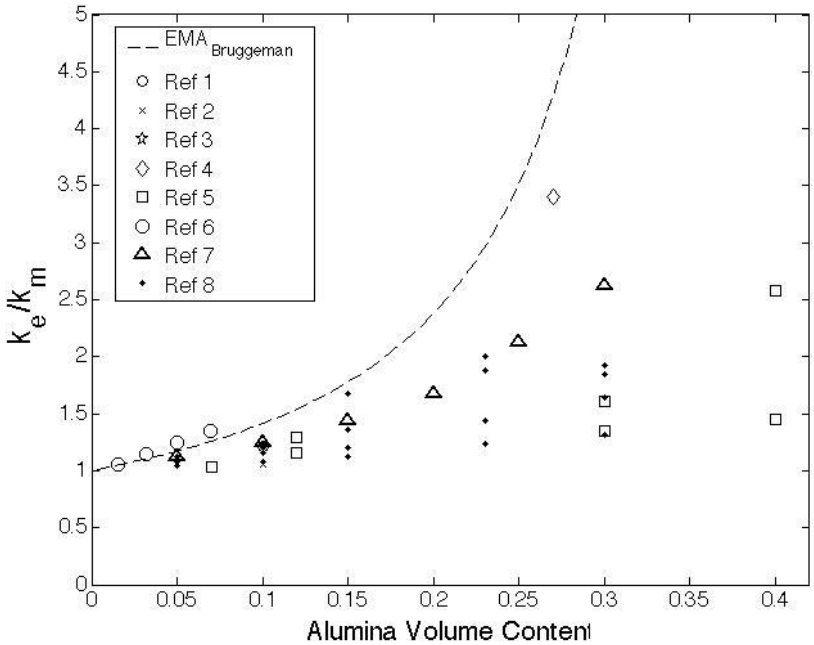
The Challenge

thermal transport in nanocomposites

Example: addition of high *k* inorganic particles to PMMA/PS
vs. effective medium approximation

Problem:
thermal boundary resistance reduces efficiency of particle fillers

Al2O3 in PMMA/PS

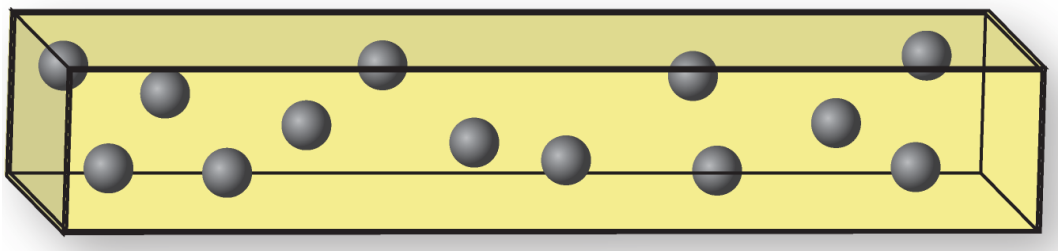


data from:

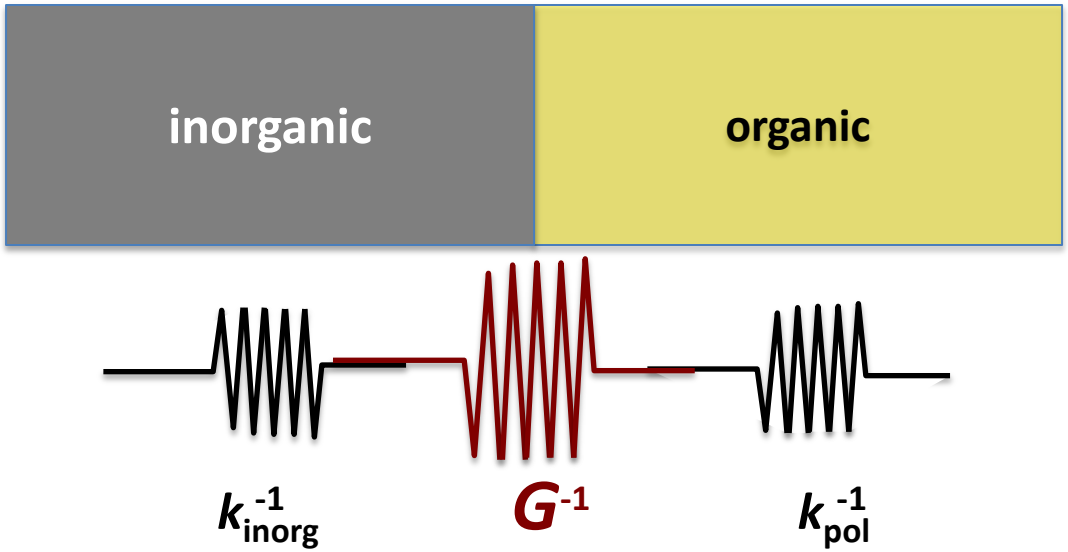
[1] Ellakwa, A.; Morsy, M.; El-Sheikh, A. Journal of Prosthodontics 2008, 17, 439–444. [2] Putnam, S.; Cahill, D. J. Appl. Phys. 2003, 94 (10) [3] Wong, T. Thermal Conductivity, Technomic Publishing, 1994. [4] Tanaka, T.; Kozako, M.; Okamoto, K. IEEE 2012, 2(1) 90–98. [5] Lu, X.; Xu, G. J. Appl. Pol. Sc.1997, 65, 2733–2738. [6] Dong, H.; Fan, L.; Wong, C.P. Electronic Components and Technology Conference 2005, 1451–1454. [7] Agari, Y.; Ueda, A.; Nacai, S. J. Appl. Poly. Sci. 1993, 49, 1625–1634. 8] Zhang S.; Cao, X.Y.; Ma, Y.; Ke, Y.C.; Zhang, J.K.; Wang, F.S. Polym. Lett. 2011, 5(7) 581–590.

The Challenge

interface boundary resistance



presence of interface gives rise to *boundary resistance* that reduces heat flow within material



for particles to raise *k* the size has to exceed a *threshold size*

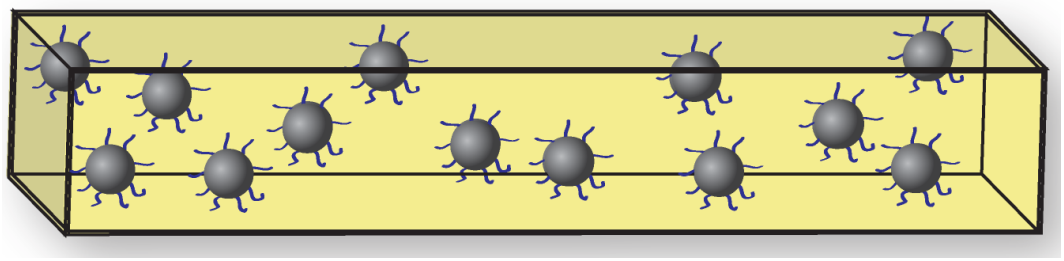
$$R > r_K \text{ (~ xx nm)}$$

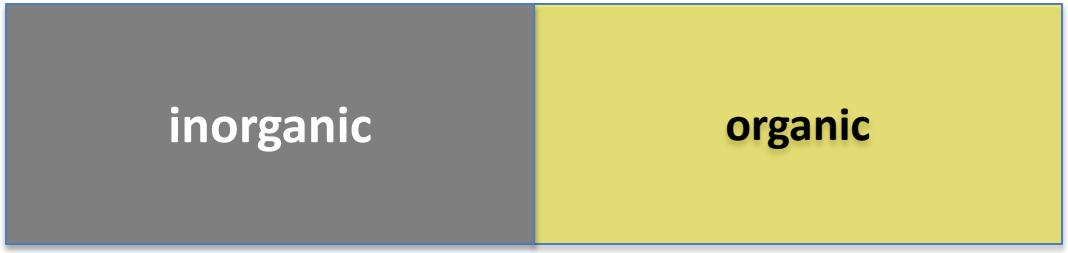
note:

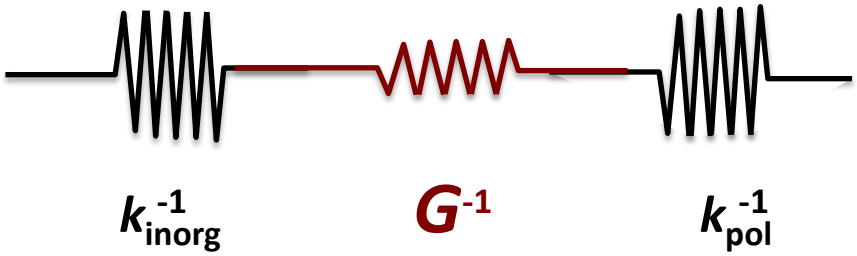
G^{-1} is sensitive to presence of ligands

The Challenge

interface boundary resistance







boundary resistance decreases with

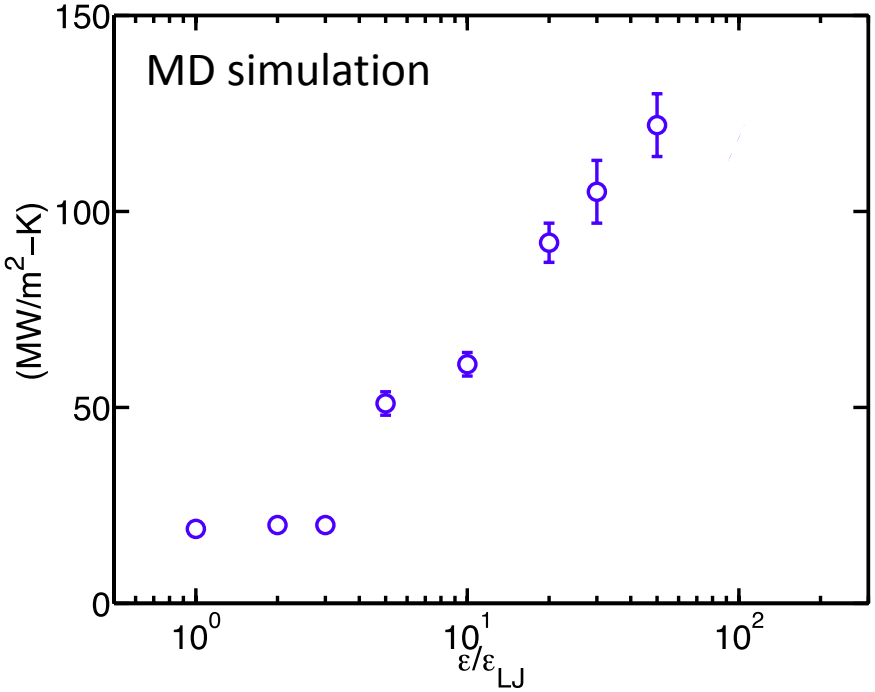
(a) increasing number of ligands

(b) increasing bonding strength of ligand/inorg/org interactions

The Challenge

role of ligand bonding strength on boundary resistance

results: **boundary conductance increases with coupling strength**



material system:

alkyl – X – silica

conclusion

use covalent tethers where possible

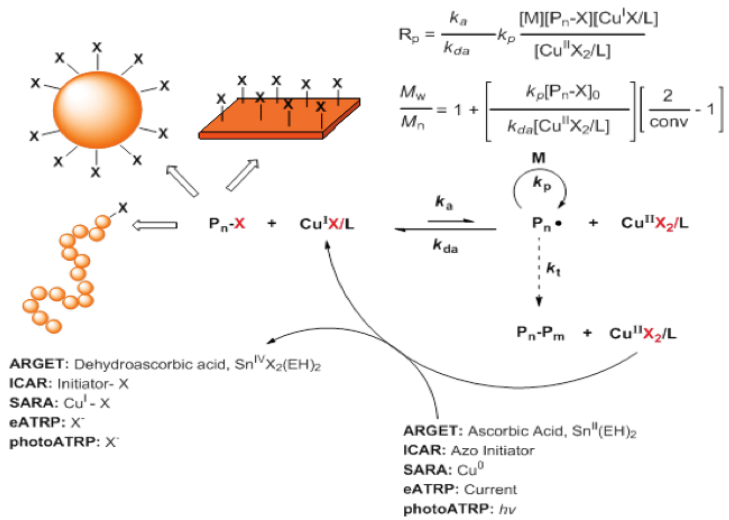
with A. McGaughey & J. Malen

informs surface modification strategies

Our Expertise

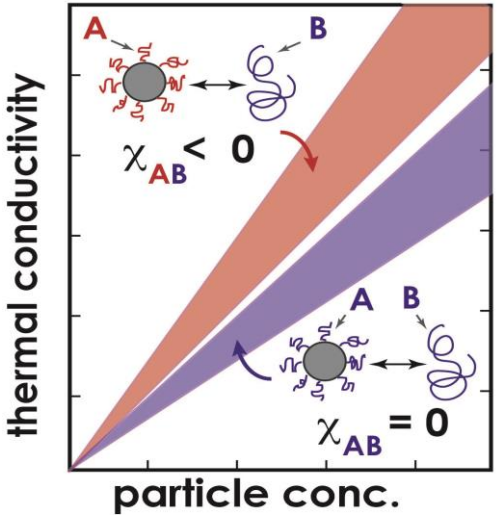
Chemistry & Physics of Polymer-Tethered Interfaces

from polymer modification …



scheme of SI-ATRP process

to transport & interactions in hybrids



raising *G* conductance by polymer-graft modification raises *k* in polymer glasses

ACS Macro Letters **2014**, 3, 1059-1063. *Nature Commun.* **2015**, 6, 8309.

Chem. Mater. **2014**, *26* , 745–762. *ACS Appl. Mater. Interfaces* **2014**, 6, 21500.

Carnegie Mellon

Title: **Novel Transparent Phosphor Conversion Matrix with High Thermal Conductivity for Next Generation Phosphor-Converted LED-based Solid State Lighting**

Agreement #: DE-EE0006702

Prime Recipient: Carnegie Mellon University

Topic Area: Area 1 – LED Core Technology Research
LED Down Converters (MYPP Task A.1.3)

Program Manager: Joel S. Chaddock

PI: Michael R. Bockstaller
Department of Materials Science and Engineering, Carnegie Mellon University, 5000 Forbes Ave., Pittsburgh, PA 15243

DOW CORNING

Carnegie Mellon

OSRAM SYLVANIA

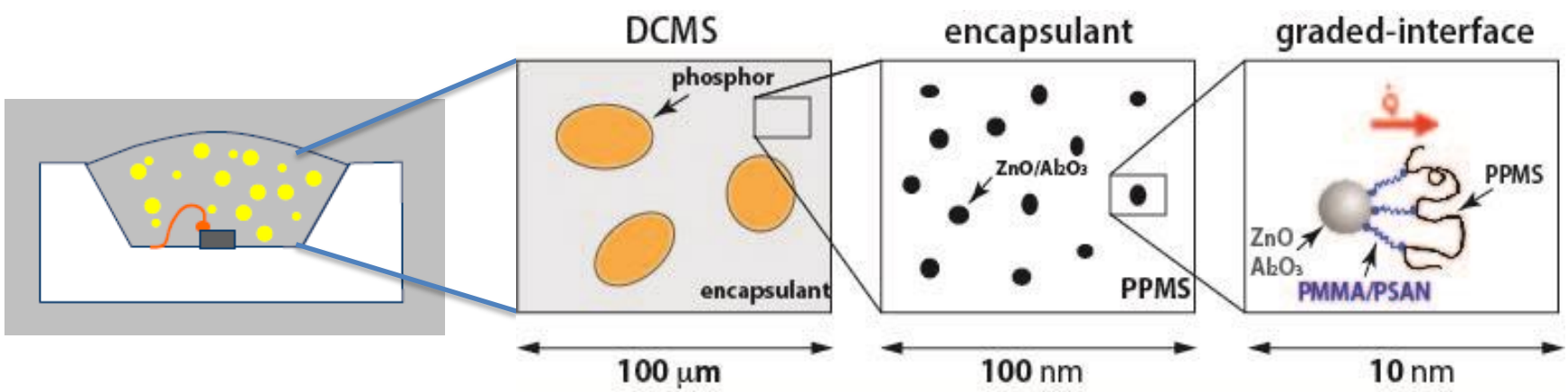
DE-EE0006702

research scope:

I. develop polymer graft enabled graded-interface modification for high *k* Al_2O_3 & ZnO particle fillers to raise thermal boundary conductance

II. develop dispersion & processing schemes for integration of fillers into commercial siloxane-based LED encapsulants, demonstrate *k* > 1 W/mK

III. evaluate device performance



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Concept

pl. visit our poster

DE-EE0006702

use surface polymerization processes to enable surface of particle fillers to facilitate:

- **increase of thermal conductance of particle/matrix interface**
  - enable higher k_{eff} at reduced inorganic loading
- **uniform dispersion of particle within matrix**
  - reduce scattering losses, improve auxiliary physical properties
- **reduction of the scattering cross section of particle fillers**
  - enable higher optical transparency at given inorganic loading

High-*k* Hybrid Encapsulants

Considerations

- **thermal conductivity**
- optical transparency
- photothermal stability
- processibility
- scalability

Conclusion

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increasing '*k*' of siloxanes for LED packages

- ✓ **high risk/high gain objective**
- ✓ **novel modification techniques show promise to improve on current limitations** but conditions need to be optimized
- ➢ **challenges remain: opt. transparency & photothermal stability**
  (what are fundamental physical barriers ???)

related high value targets:

increasing refractive index without compromising auxiliary phys. prop. (a more worthwhile near-term goal ?!)

new (enhanced) siloxane chemistries/molecular approaches