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Ultra-Low Thermal Conductivity of Polyhedral Oligomeric Silsesquioxanes (POSS)

Daniel Drury[1], John Tomko[2], Jeff Braun[2], Erika Vreeland[3], Patrick E. Hopkins[2], Paul G. Clem[1]*, Stanley S. Chou[1]*

1. Department of Electronic, Optical and Nanomaterials, Sandia National Laboratories, Albuquerque, NM 87185, USA
2. Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA
3. IR Dynamics LLC, 6901 Gruber NE, Suite G, Albuquerque, NM 87109

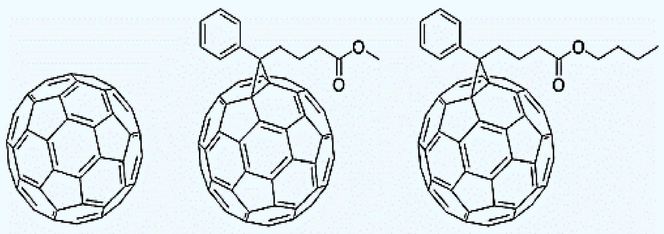
* schou@sandia.gov, pgclem@sandia.gov

Motivation: ARPA-E SHIELD (Single-Pane Highly Insulating Efficient Lucid Designs)

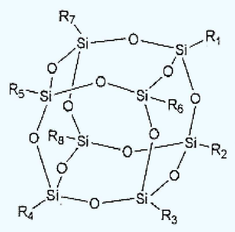
- Develop a low thermal conductivity material that is visibly transparent for application to single-pane windows to mimic the energy efficiency of double-pane windows
  - > 60% of commercial and residential heat is lost through only 20% of the building's façade through inefficient windows
  - SHIELD aims to improve the thermal insulation of single-pane windows by >10X through a simple to affix add-on
  - Potential to reduce the equivalent energy consumption of up to 32 million homes in the US

Background

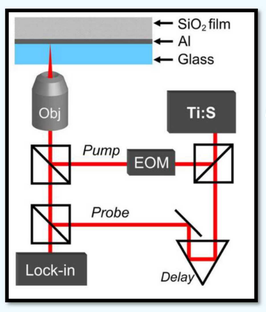
- **Lowest thermal conductivity for a fully dense film = 0.03 – 0.06 W/m·K** [1]
  - [6,6]-phenyl C 61-butyric acid methyl ester (PCBM) [6]
  - The fullerene derivative exhibited a functional group dependence
  - Drawbacks: visibly opaque and difficult to chemically derivatize
- Cage-like materials (i.e. clathrates) possess highly anharmonic frequency vibrations which lead to enhanced phonon scattering [5]
- **POSS structure: Si_8O_{12} cage with configurable functional groups**
  - Si-O bonding leads to high transparency in the visible range
  - Easily chemically modified for optical and solution behavior
  - Solubility leads to solution processable methods (screen printing, spraying)
  - Commercially available product allows for manufacturing scalability



C60 PCBM PCBNB

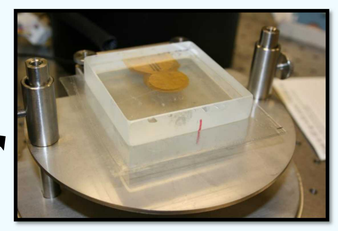


POSS cage with functional groups at the corners



Thermal Property Measurements

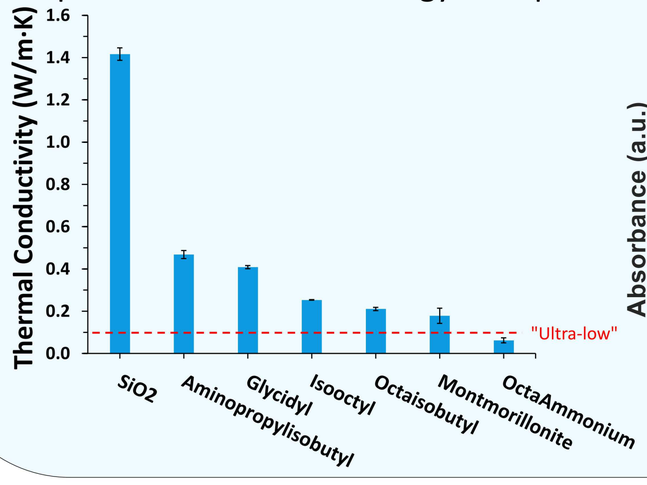
- Time-domain thermoreflectance (TDTR)[4]
  - Measures temperature-dependent reflectance of metal-coated sample
- Hot Disk Transient Plane Source (TPS)
  - Sensor acts as a heat source and thermometer
  - Average sensor temperature increase expression [2]



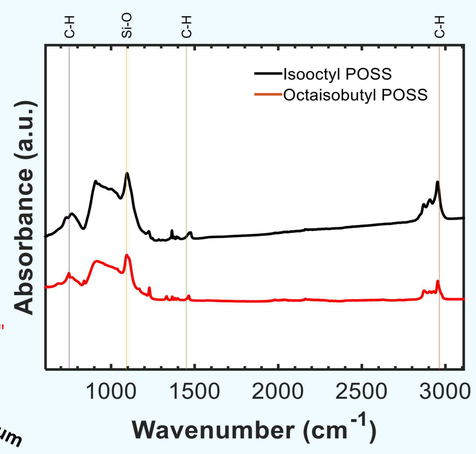
$$\overline{\Delta T(\tau)} = \Delta T_i + \frac{P_o}{\pi^{\frac{3}{2}} a \Lambda} D(\tau)$$

Results

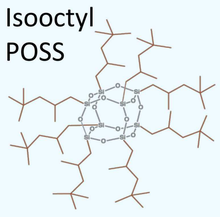
- Thermal conductivity of POSS varies between 0.06 – 0.45 W m^{-1} K^{-1} due to configurable chemical moieties
- Chemical functionalities have a strong impact on the thermal energy transport



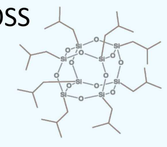
- Fourier-transformer infrared spectroscopy (FTIR) distinguishes between similarly structured POSS molecules
- Normalized absorption for Isooctyl POSS exhibits higher C-H bonding signal than Octaisobutyl POSS

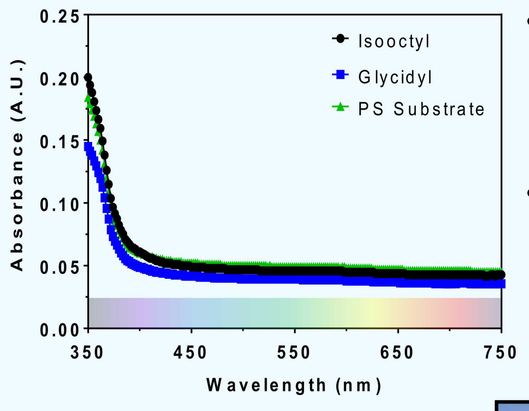


Isooctyl POSS



Octaisobutyl POSS



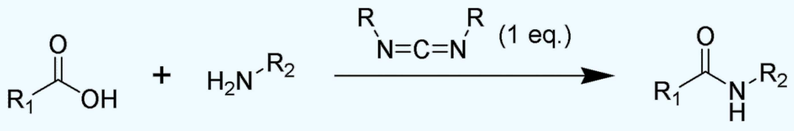


- UV-vis spectroscopy of POSS confirms that the 1 mm thick samples are visibly transparent
- Similar absorption spectrum as polystyrene (PS) of the same thickness

Construction of 1 mm thick Isooctyl POSS layer between two glass slides

Future Work

- Conjugate POSS moieties via carbodiimide and EDC reactions to correlate thermal conductivities with branch structure[3]



- Study how changes in chemical functionalization of the Si_8O_{12} cage in solution affects the low frequency thermo-coupling of the molecule and solvent
- Investigate how scaling from bulk to thin film impacts thermal properties
- Integrate with thermochromic V_2O_3 nanoparticles to reflect/transmit heat

Conclusion

Found a strong contender to meet the SHIELD requirements set by ARPA-E

- Ultra-low thermal conductivity (0.06 – 0.45 W/mK)
- Visibly transparent films with no noticeable haze
- Applicable to variety of substrates

Acknowledgements

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