Designing Advanced Thermoelectric Materials for Automotive Applications

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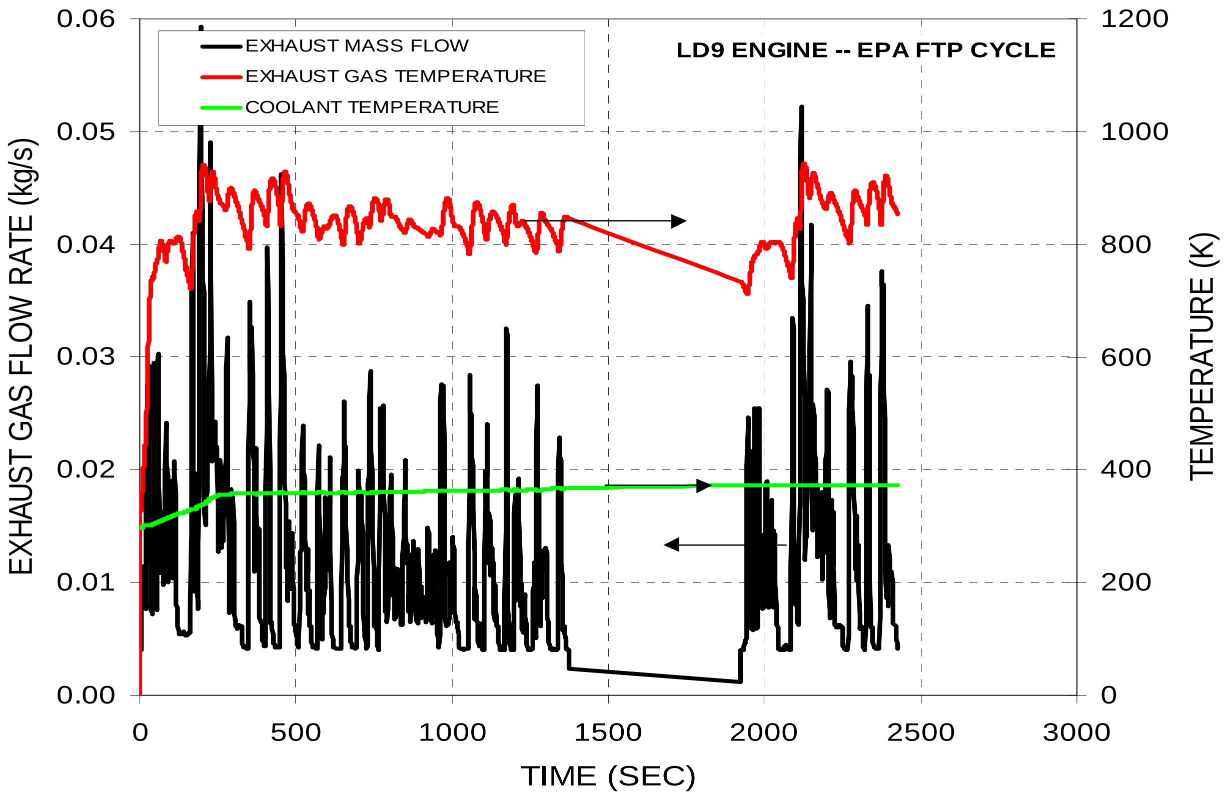
Outline

- Automotive relevance
- ZT requirements
- Recent material advances
- Our study on $CoSb_3$ with fullerene additions
- Conclusions

Exhaust Flow and Temperatures for a 4- Cylinder Engine



- Tens of kW of waste heat energy in the exhaust and coolant

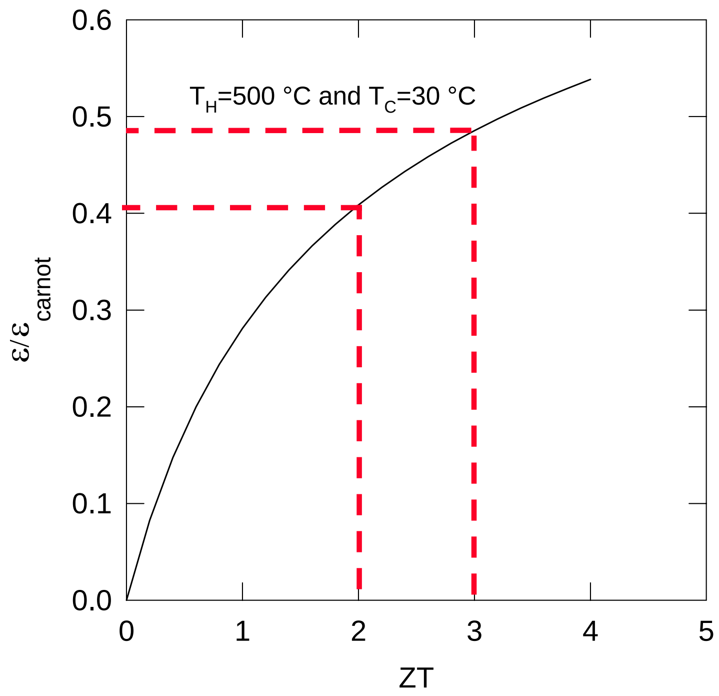
Environmental Impact of Mobile AC Refrigerant: Global Warming

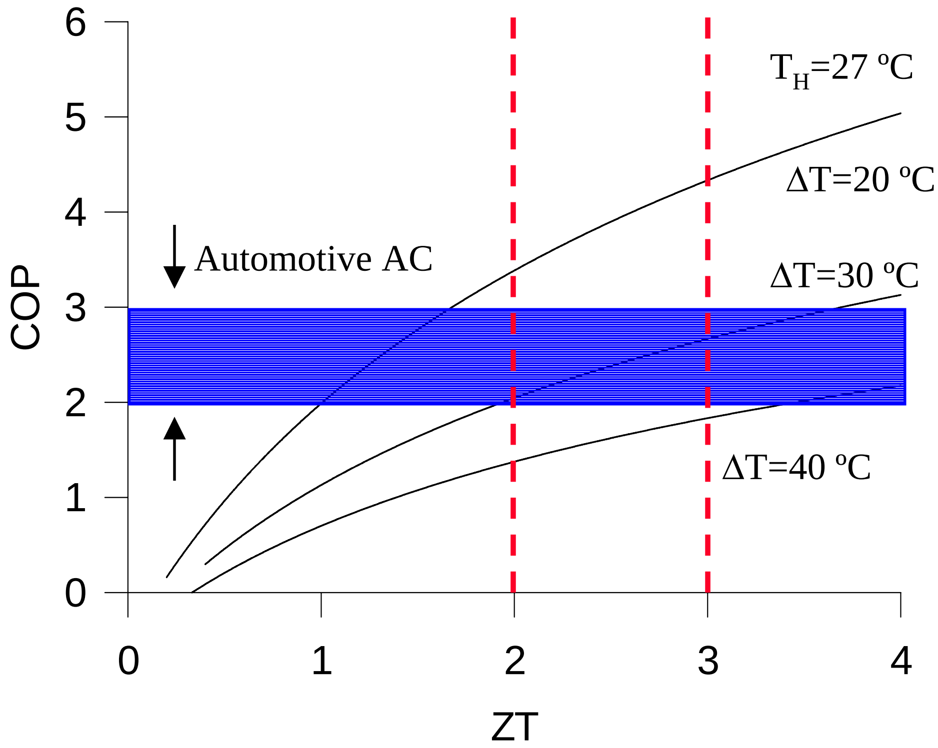
- Refrigerant leakage
- Increase in emission due to AC
- Thermoelectric cooling could be potential long range climate control technology for automobiles

TE Power Generators & Coolers (Solid State Heat Pumps)

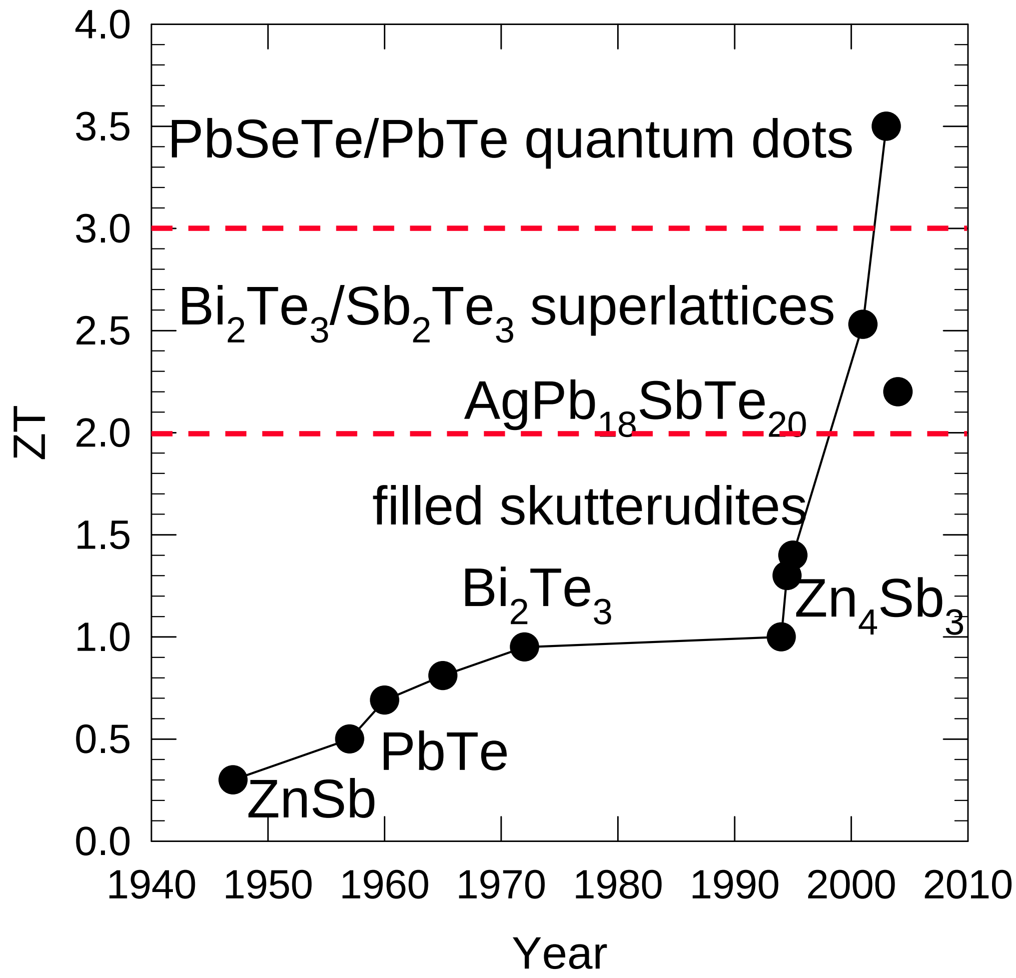
- Advantage:
  - All solid state operation
  - Easily controllable electronically
  - Complete reversible for heating and cooling
  - Low noise and vibration, extremely reliable
  - No harmful refrigerants
- Disadvantage:
  - Low energy conversion efficiency
  - Low COP

TE Power Generation & Refrigeration

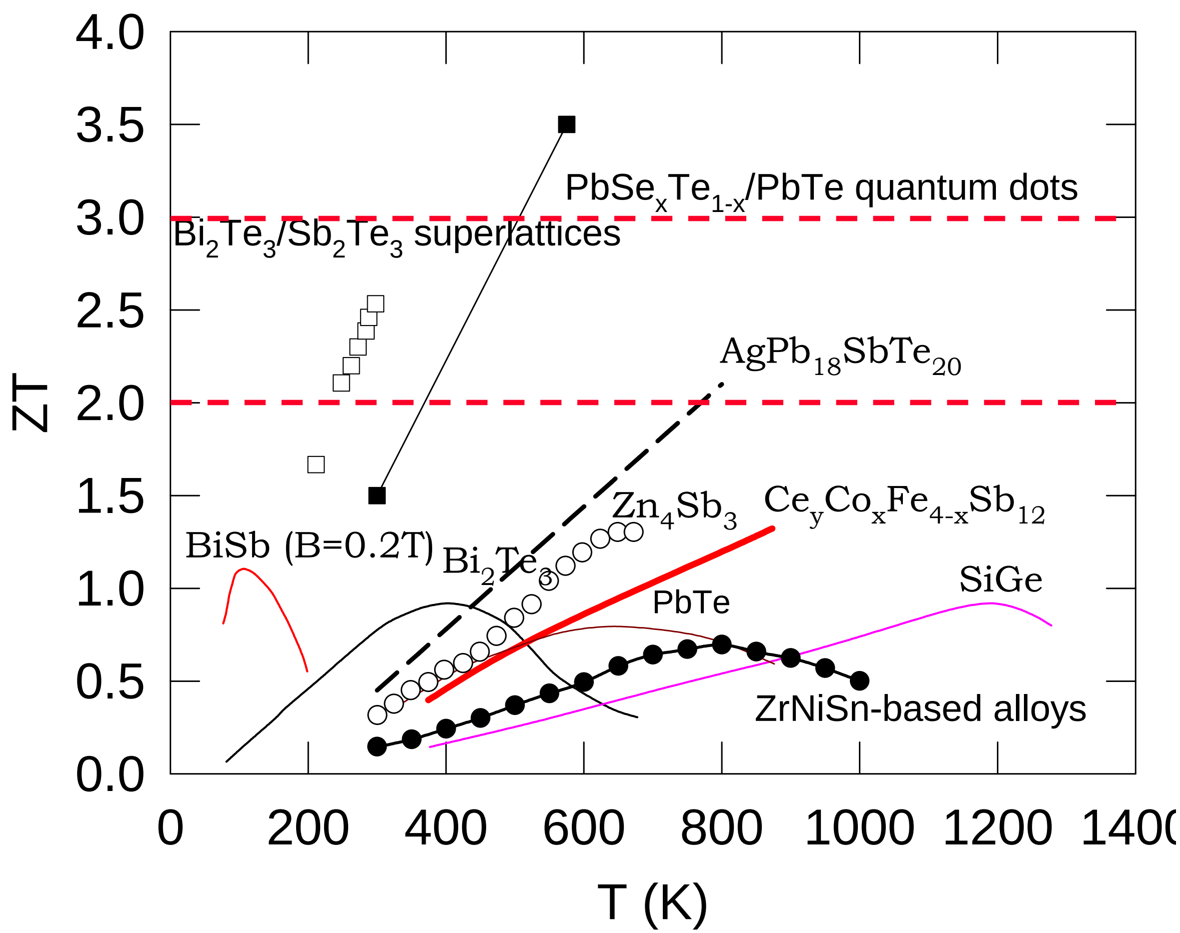




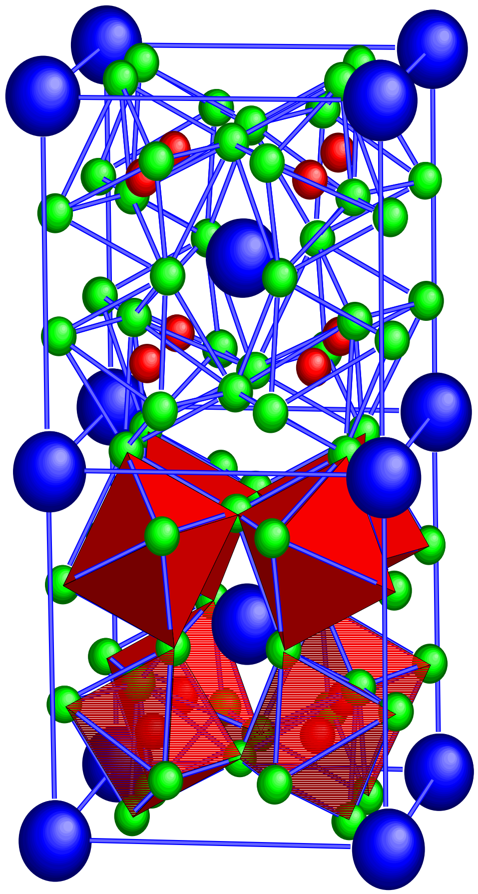
Timeline of ZT



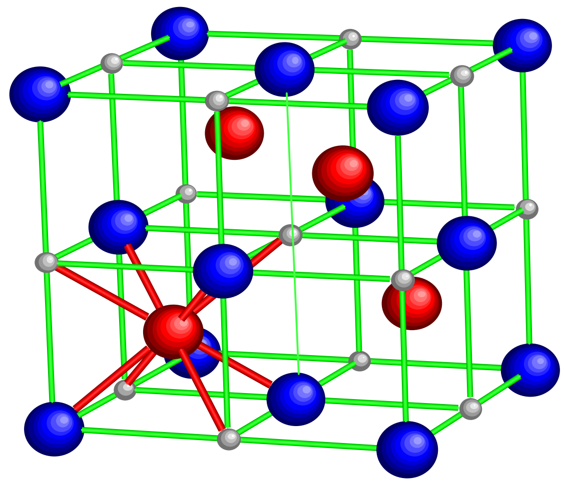
Temperature Dependence of ZT



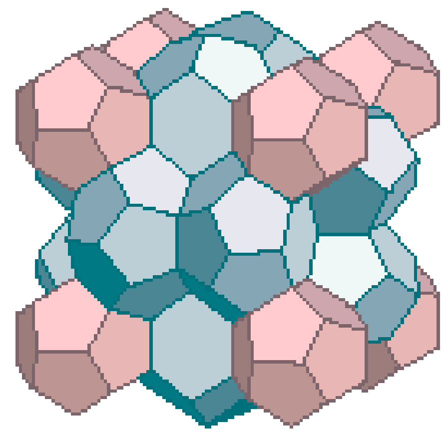
Material Systems Studied



Skutterudites



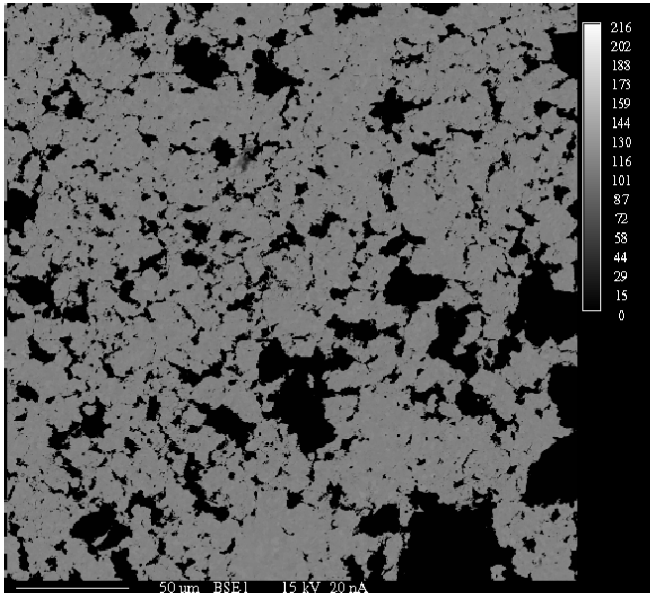
Half-Heuslers



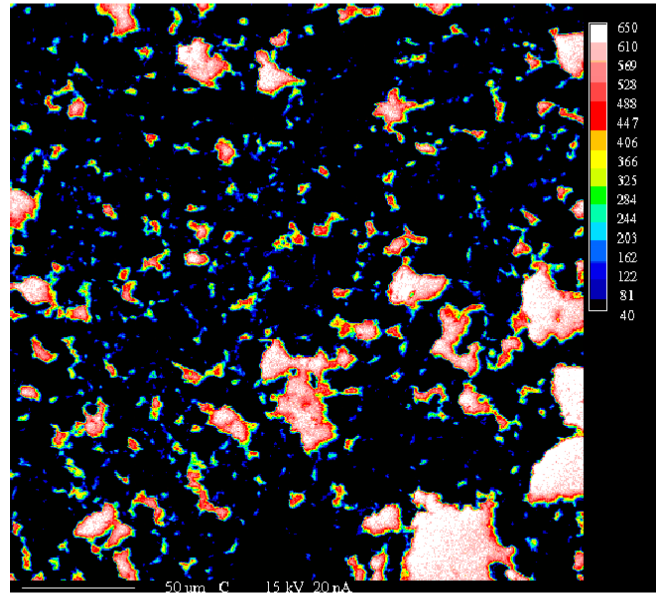
Clathrates

- Elucidate the crucial factors in these compounds relating to various sublattices, particularly with respect to compositions, electronic doping levels, and phonon scattering mechanisms, and hence optimize their TE properties

$CoSb_3$ + C_{60} Composites



BSE



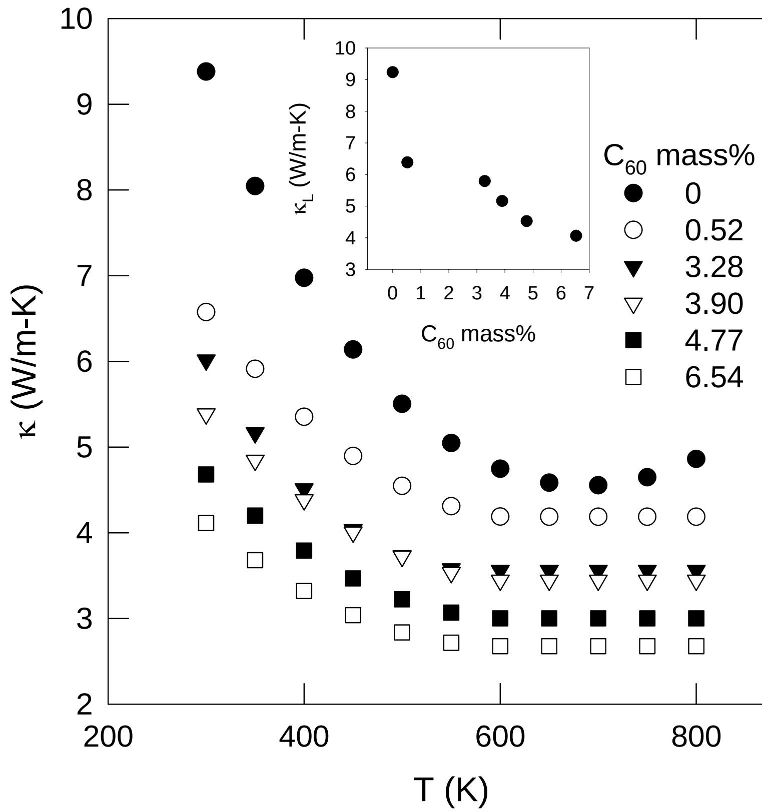
Carbon x-ray map

- C_{60} molecules agglomerate into irregular micro-sized clusters at the grain boundary
- Cluster sizes vary from a few to 50 µm
- Average cluster size increases with increasing C_{60} content

C_{60} Content, Skutterudites Phase Composition, and Hall Data

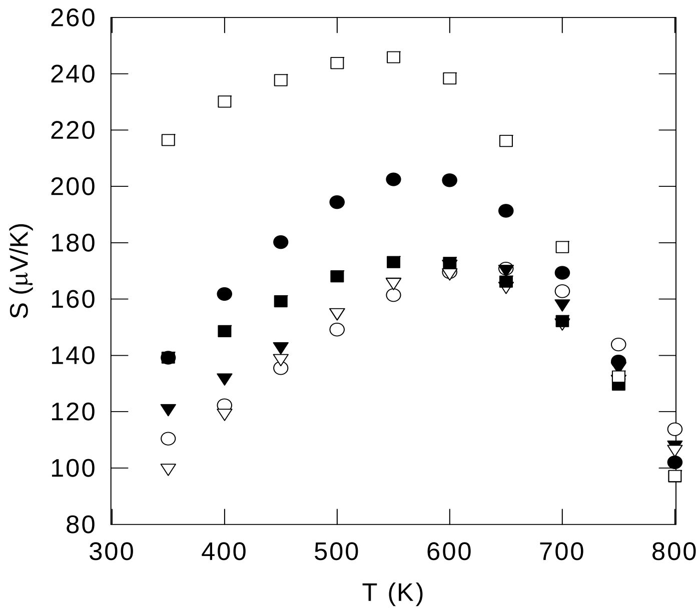
Nominal C_{60} mass%	Actual C_{60} mass%	Chemical composition of the skutterudite phase	Room temperature hole concentration (10^{18} cm^3)
0	0	$CoSb_{2.995}$	1.30
2	0.52	$CoSb_{3.008}$	5.03
4	3.28	$CoSb_{3.004}$	4.67
5	3.90	$CoSb_{3.004}$	5.76
6	4.77	$CoSb_{3.003}$	3.75
8	6.54	$CoSb_{2.998}$	1.38

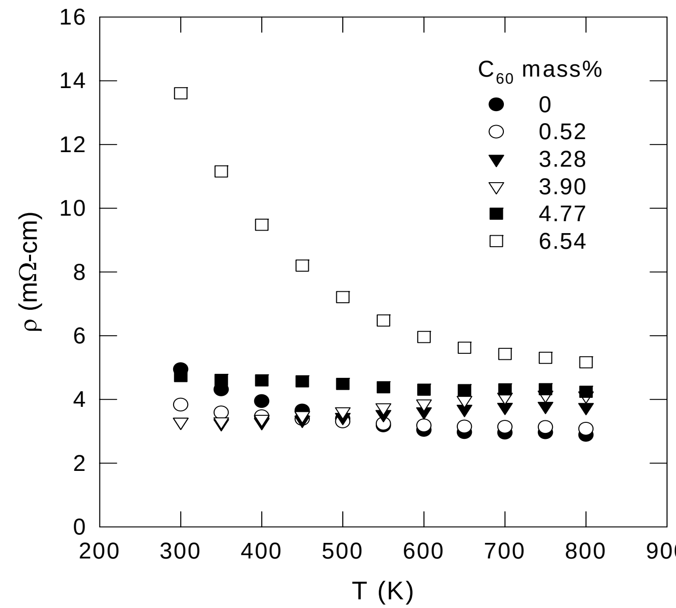
Thermal Conductivity



- κ = 9.38 W/m-K for $CoSb_3$ at room temperature, agrees with the literature data
- κ decreases with increasing C_{60} content
- Phonon wavelength $\lambda_p << L_{clusters}$, κ reduction is due to phonon-large defect scattering
- This type of phonon scattering depends on the size and content of the defects, but is independent of ϖ_p

Thermopower & Resistivity



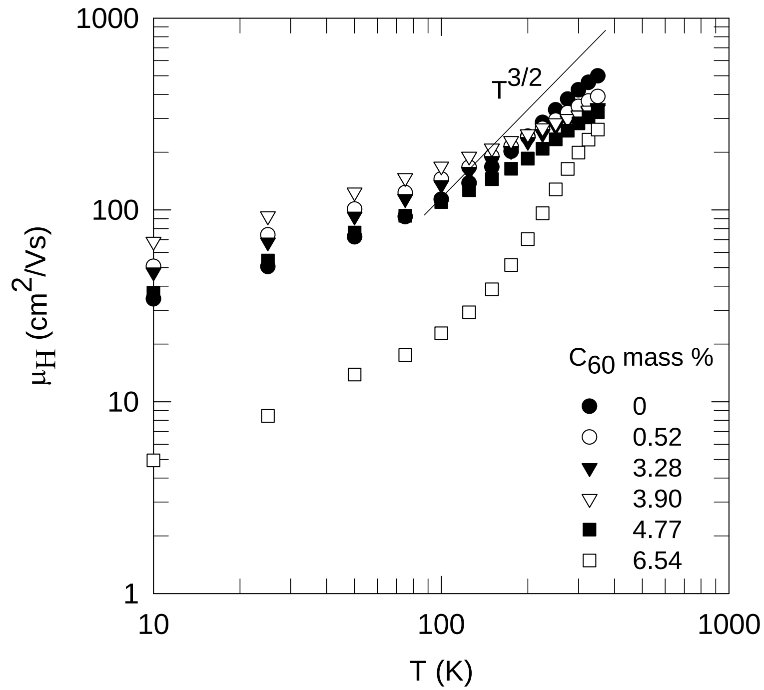


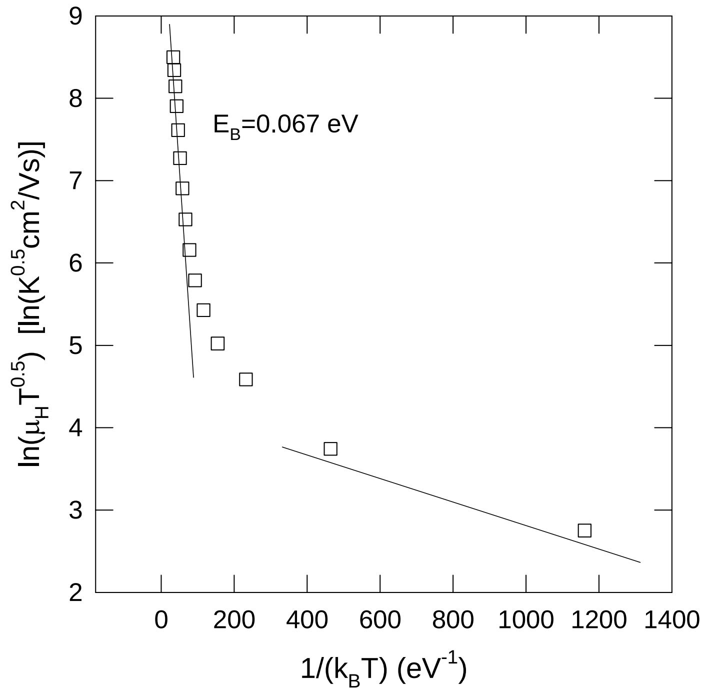
- At room temperature, S and ρ variations for samples with < 5 mass% C_{60} are due to carrier concentration variations observed
- The much enhanced S and ρ for $CoSb_3$ with 6.54 mass% C_{60} reflect a carrier scattering mechanism change because

$$S = \mp \frac{k_B}{e}\left[\eta_F - \frac{(r+\frac{5}{2})F_{r+3/2}}{(r+\frac{3}{2})F_{r+1/2}}\right]$$

$$\rho = ne\mu$$

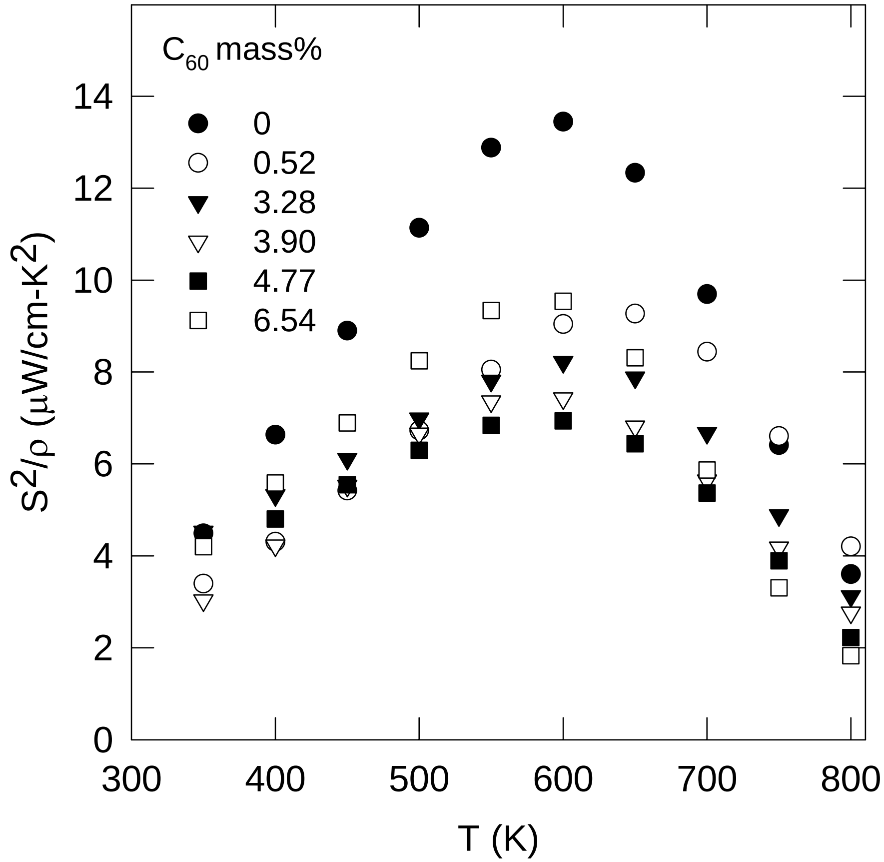
Hall Mobility





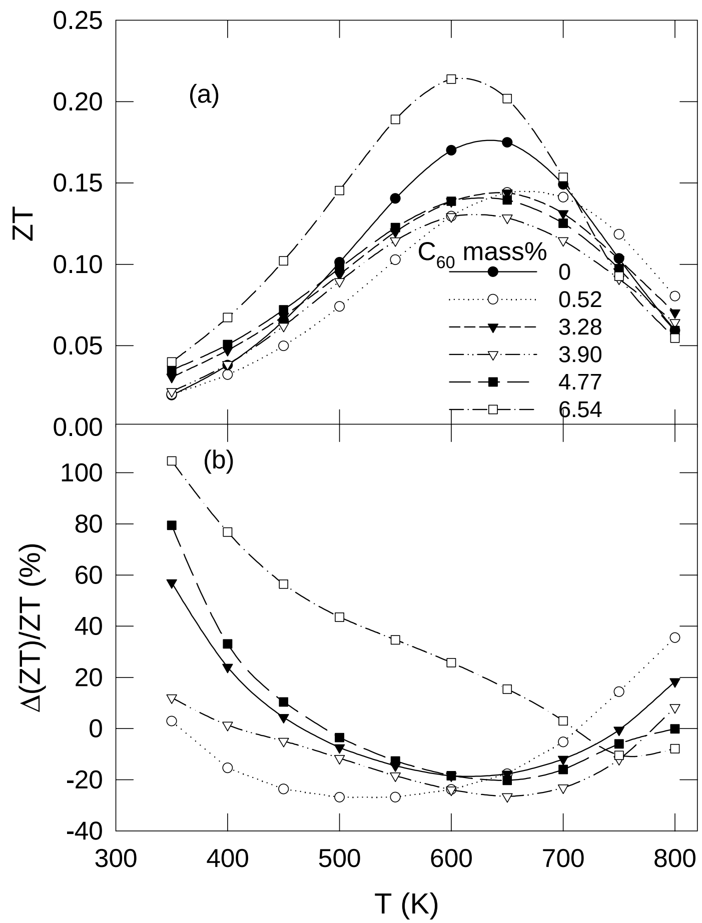
- For hole ionized impurity scattering: $\mu_H \propto T^{3/2}$
- For hole grain barrier scattering: $\mu_H = \dfrac{el}{\sqrt{8k_BT\pi m^*}} e^{\frac{-E_B}{k_BT}}$
- A transition from ionized impurity scattering to grain barrier scattering dominated electrical transport occurs ~ 5 mass% C_{60}

Power Factor



- Power factor reduction for $CoSb_3$ with 6.54 mass% C_{60} is only modest

ZT



- Significant *ZT* increase for $CoSb_3$ with 6.54 mass% C_{60}, due to large κ decrease and modest power factor decrease

Conclusions

- Recent advances in thermoelectric materials research show promise of TE technologies for automotive applications: power generation and cooling
- Our study of $CoSb_3$ with C_{60} additions show enhancement of *ZT*, especially with 6.54 mass% C_{60} addition
- Our data suggest even larger *ZT* values would be attainable for the prospective high performance TE materials such as filled skutterudites, clathrates, and half-Heuslers, by this method of C_{60} additions