Advanced Radioisotope Power Systems
Segmented Thermoelectric Research

Direct Energy Conversion Workshop
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Presented by

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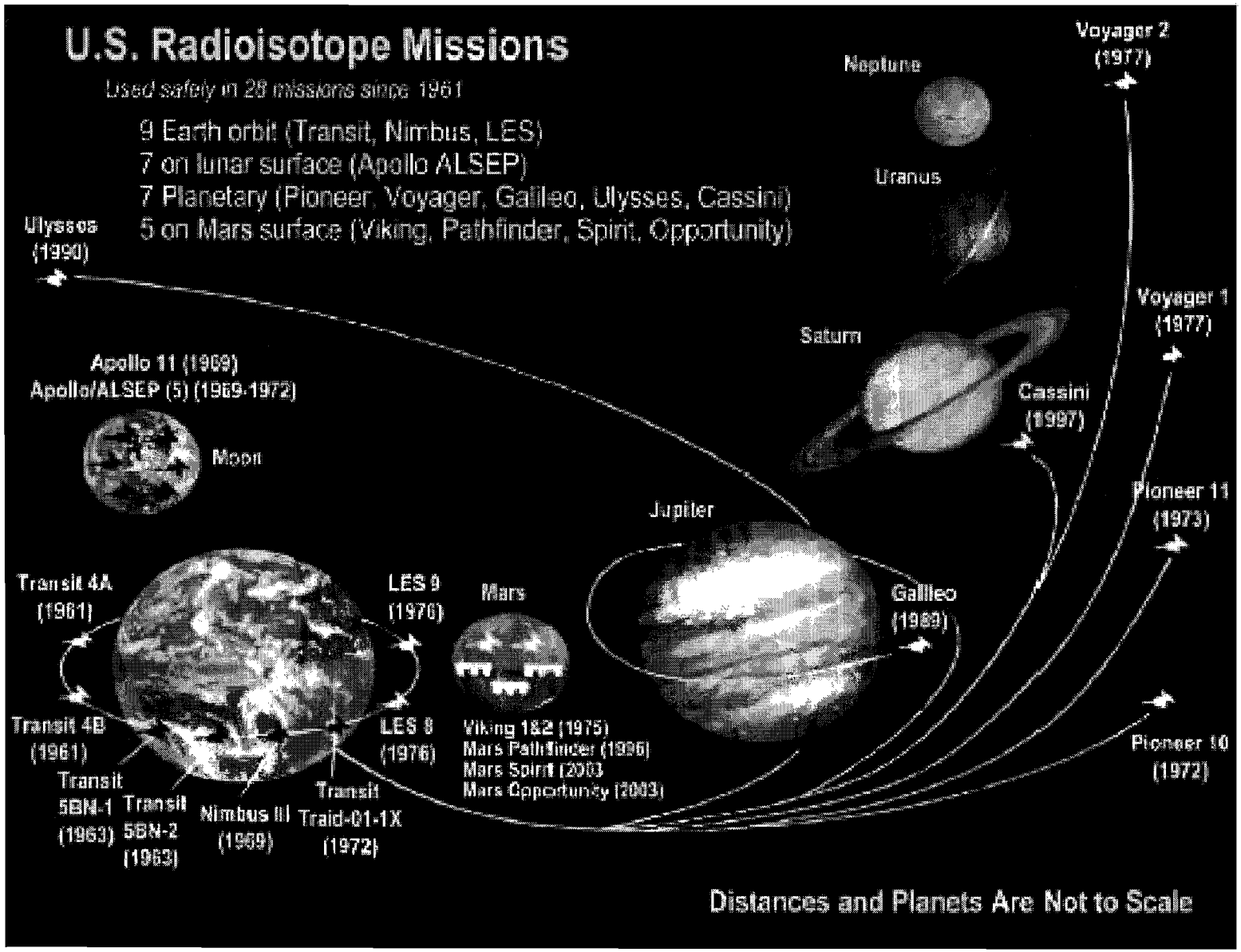
Jet Propulsion Laboratory/California Institute of Technology

Acknowledgements:

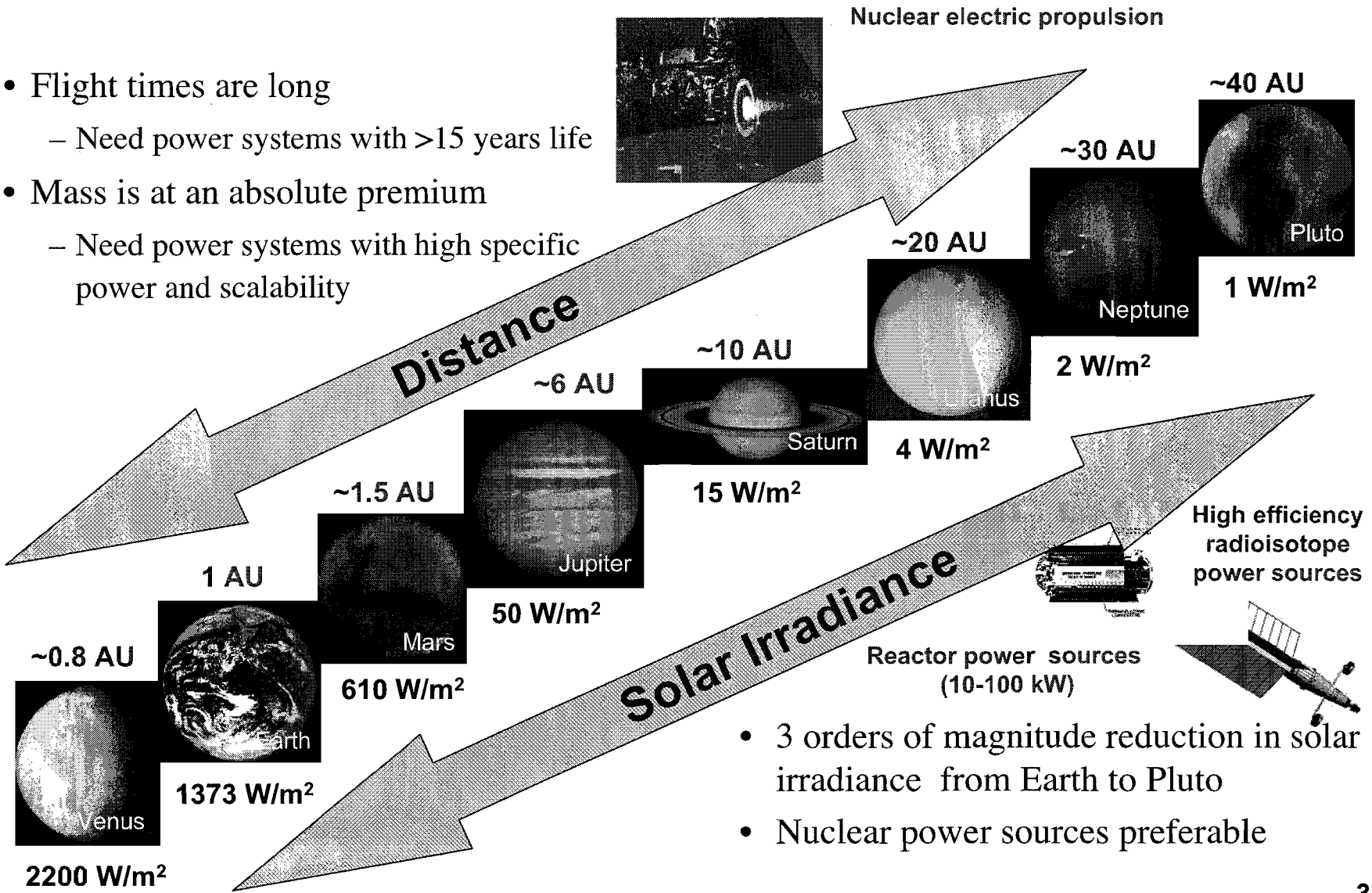
NASA, ONR & DARPA

U.S. missions using radioisotopes power and/or heating sources



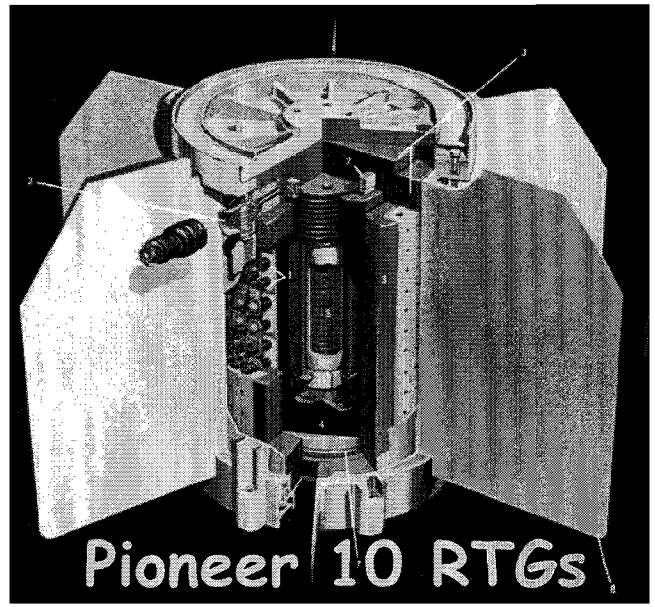
Power Technology

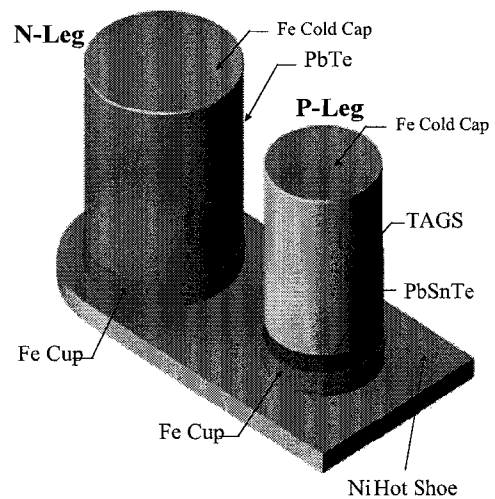
- Flight times are long
  - Need power systems with >15 years life
- Mass is at an absolute premium
  - Need power systems with high specific power and scalability



- 3 orders of magnitude reduction in solar irradiance from Earth to Pluto
- Nuclear power sources preferable

PbTe/TAGS-based systems



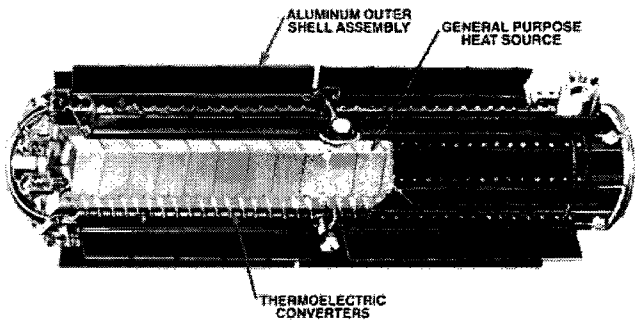


PbTe/TAG unicouples

General Purpose Heat Source RTG

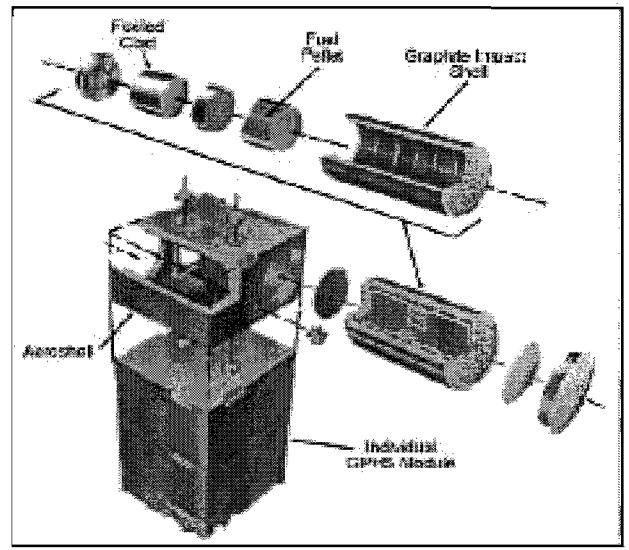
General Purpose Heat Source (GPHS)

Radioisotope Thermoelectric Generator (RTG)

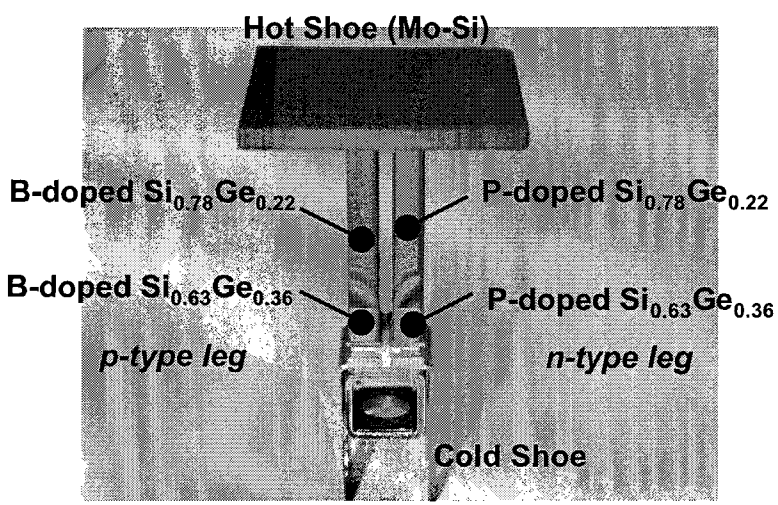


- POWER OUTPUT – 285 W(e)
- FUEL LOADING – 4400 W(t); 132,500 Ci
- WEIGHT – 124 lbs
- SIZE – 16.6 in x 44.5 in

The three Radioisotope Thermoelectric Generators (RTGs) provide electrical power for Cassini's instruments and computers. They are being provided by the U.S. Department of Energy.



General Purpose Heat Source Module



GPHS SiGe unicouple

GPHS-RTG Performance Data

Power output-We	290 beginning of life 250 end of life
Operational life - hrs	40,000 after launch
Weight-kg	55.5
Output voltage	28
Dimensions	42.2 diameter 114 long
Hot junction temperature-K	1270
Cold junction temperature-K	566
Fuel	PuO_2
Thermoelectric material	SiGe
Numbers of unicouples	572
Mass of Pu-238-g	7,561

Advanced Radioisotope Power Systems (APRS) for NASA missions

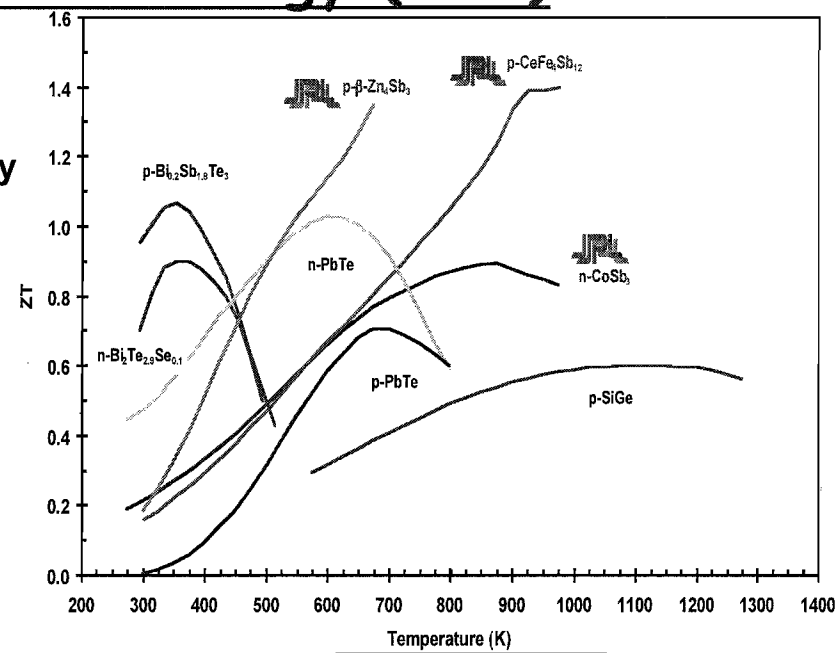
- Overall objective:

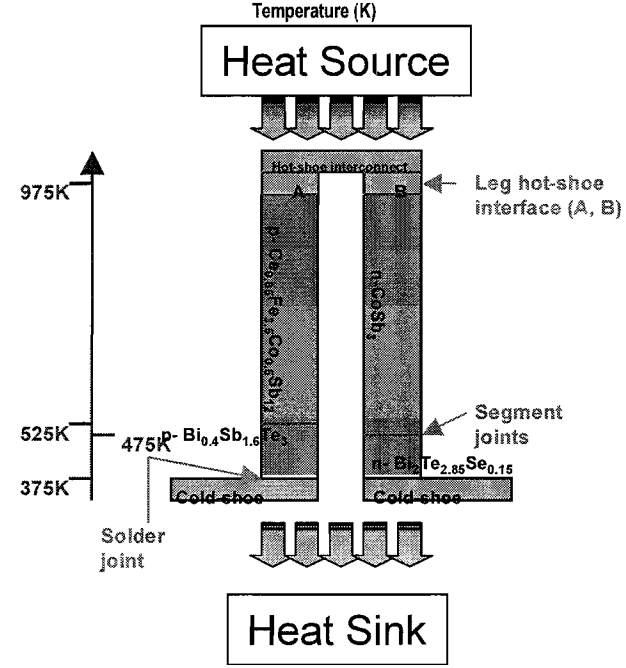
  Develop low mass, high efficiency, low-cost Advanced Radioisotope Power System with double the Specific Power and Efficiency over state-of-the-art Radioisotope Thermoelectric Generators (RTGs)

Segmented Thermoelectric Technology (STE)

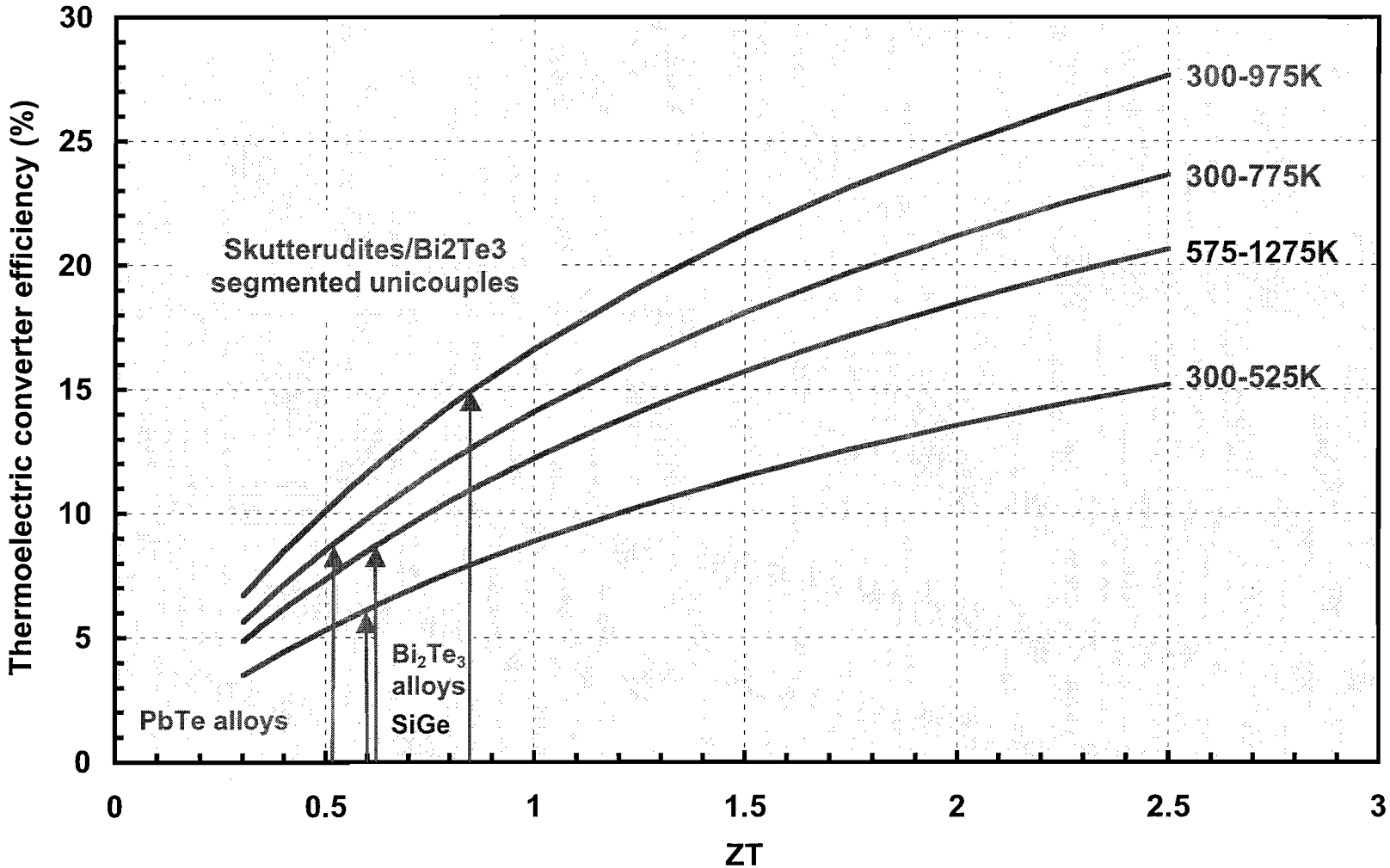
- **New high ZT materials**
  - Development initiated in 1991 and supported by ONR and DARPA
  - Higher efficiency values
- **Segmented unicouples**
  - Large ΔT, high ZT -> high efficiency
  - Using a combination of state-of-the-art TE materials (Bi_2Te_3-based materials) and new, high ZT materials developed at JPL
    - Skutterudites : $CeFe_4Sb_{12}$ and $CoSb_3$
    - Zn_4Sb_3
  - Current materials operation limited to ~ 975K
  - Higher average ZT values
  - ➔ Higher material conversion efficiency
    - ➔ Up to 15 % for a 300-975K temperature gradient

$$\text{Efficiency} \quad \eta = \frac{T_H - T_C}{T_H} \frac{\sqrt{1+ZT}-1}{\sqrt{1+ZT}+\frac{T_C}{T_H}}$$



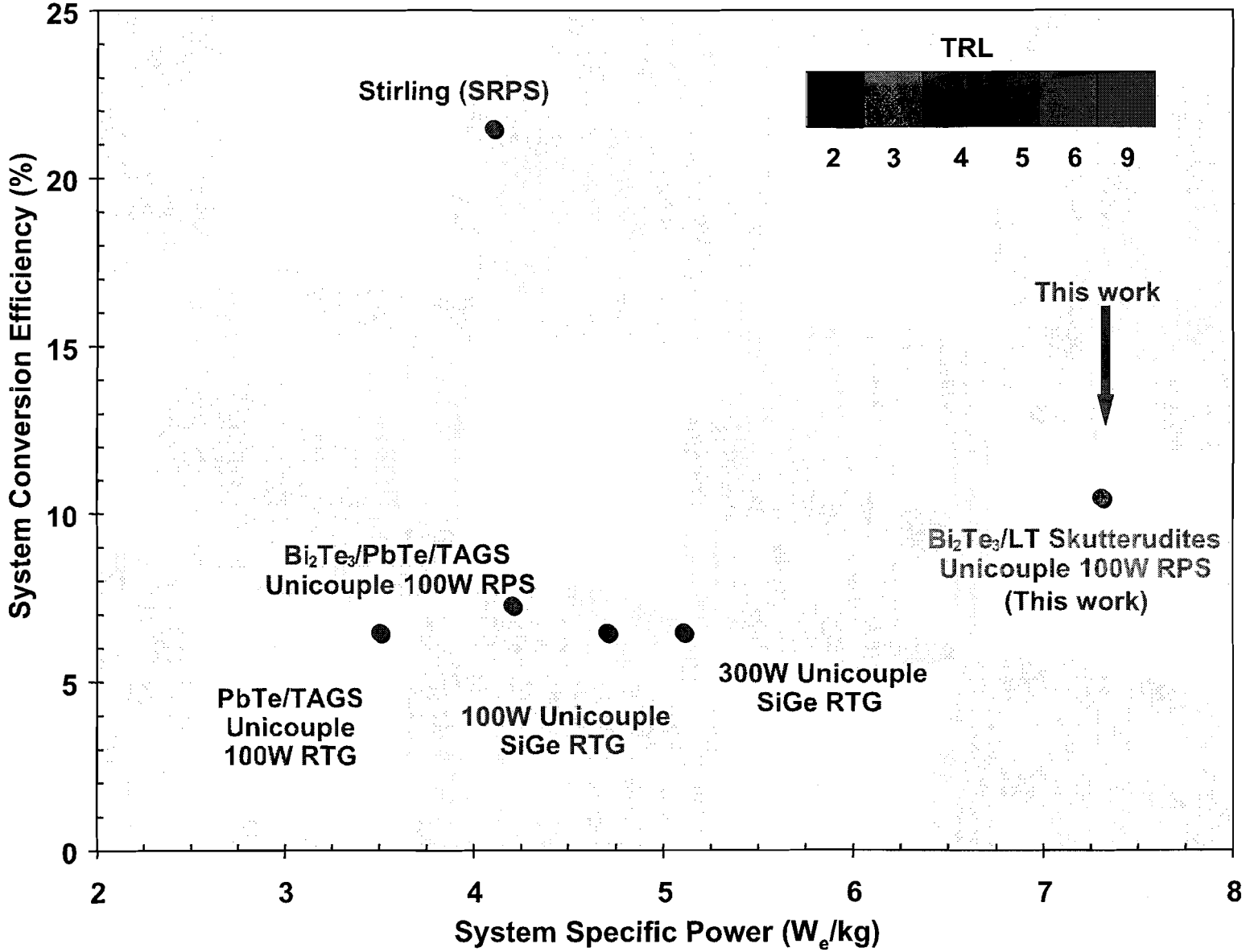


Converter efficiency : state-of-the-art vs. segmented thermoelectric technology



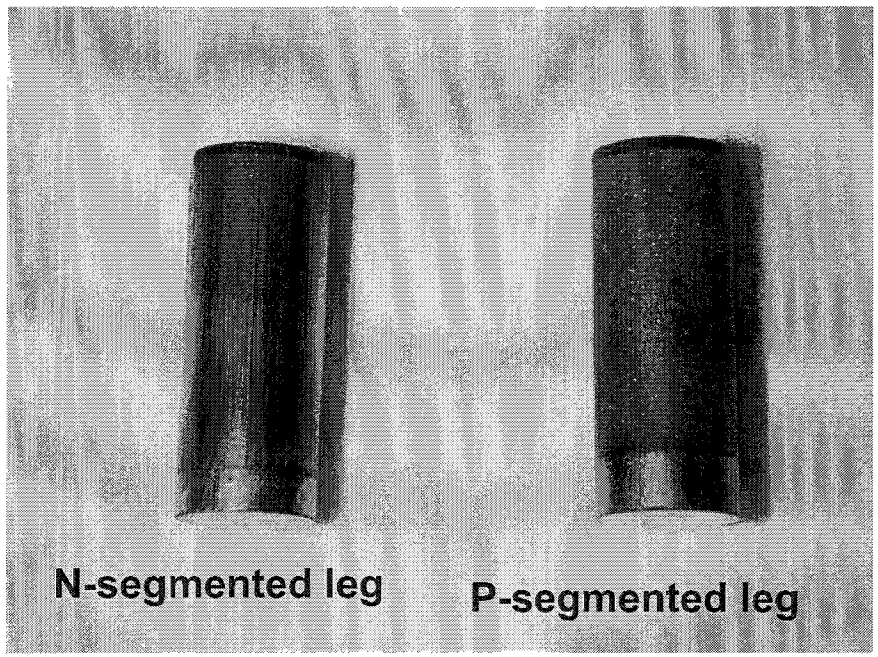
STE technology has the potential for achieving twice the converter efficiency of SOA thermoelectrics

System Efficiency & Specific Power: STE vs. SOA



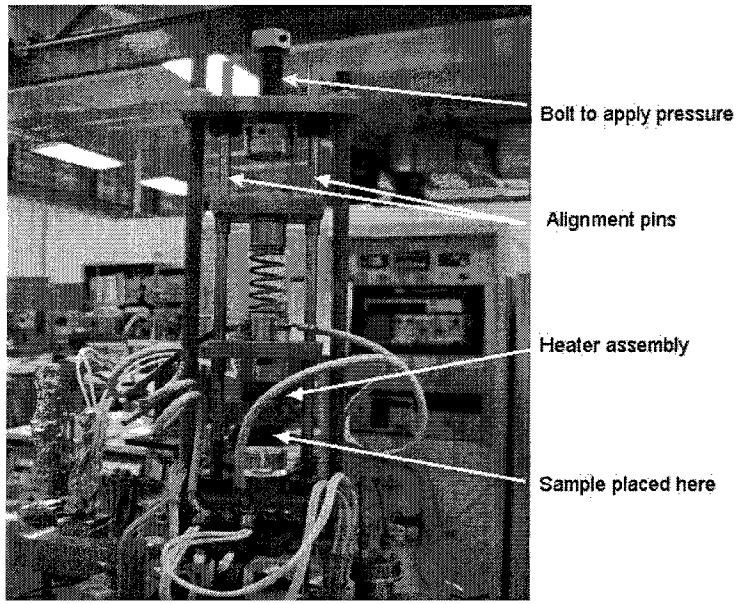
Legs metallization summary

- **P-leg layering**
  - Ti/Co/p-type $Ce_1Fe_3Ru_1Sb_{12}$/Co/Ti/p-type $Bi_{0.4}Sb_{1.6}Te_3$/Ni
- **N-leg layering**
  - Ti/n-type Pd & Te doped $CoSb_3$/Ti/p-type $Bi_2Te_{2.95}Te_{0.05}$/Ti/Ni

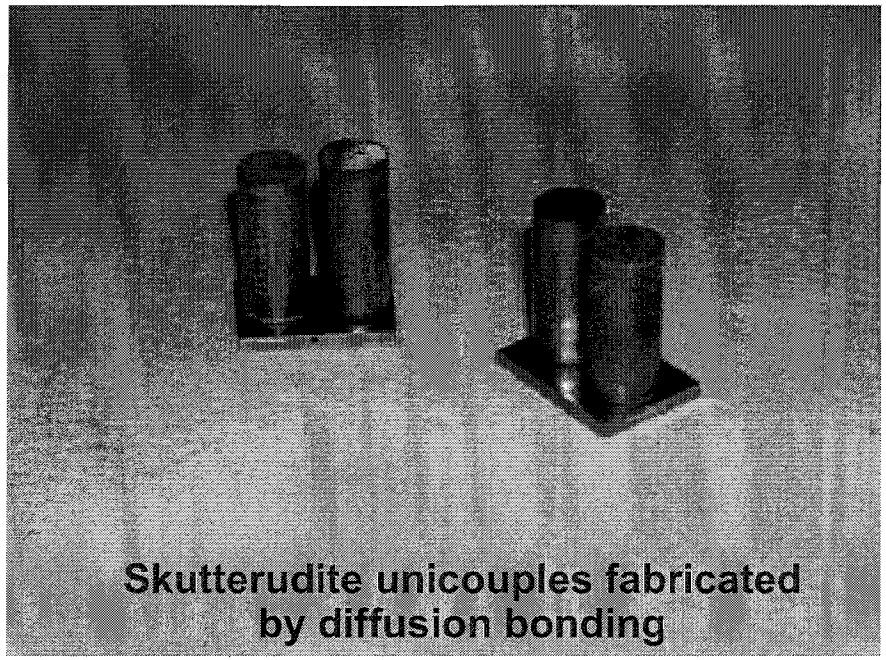


Segmented legs with metallization compatible with a Mo hot-shoe have been developed

Unicouple fabrication



Diffusion bonding station

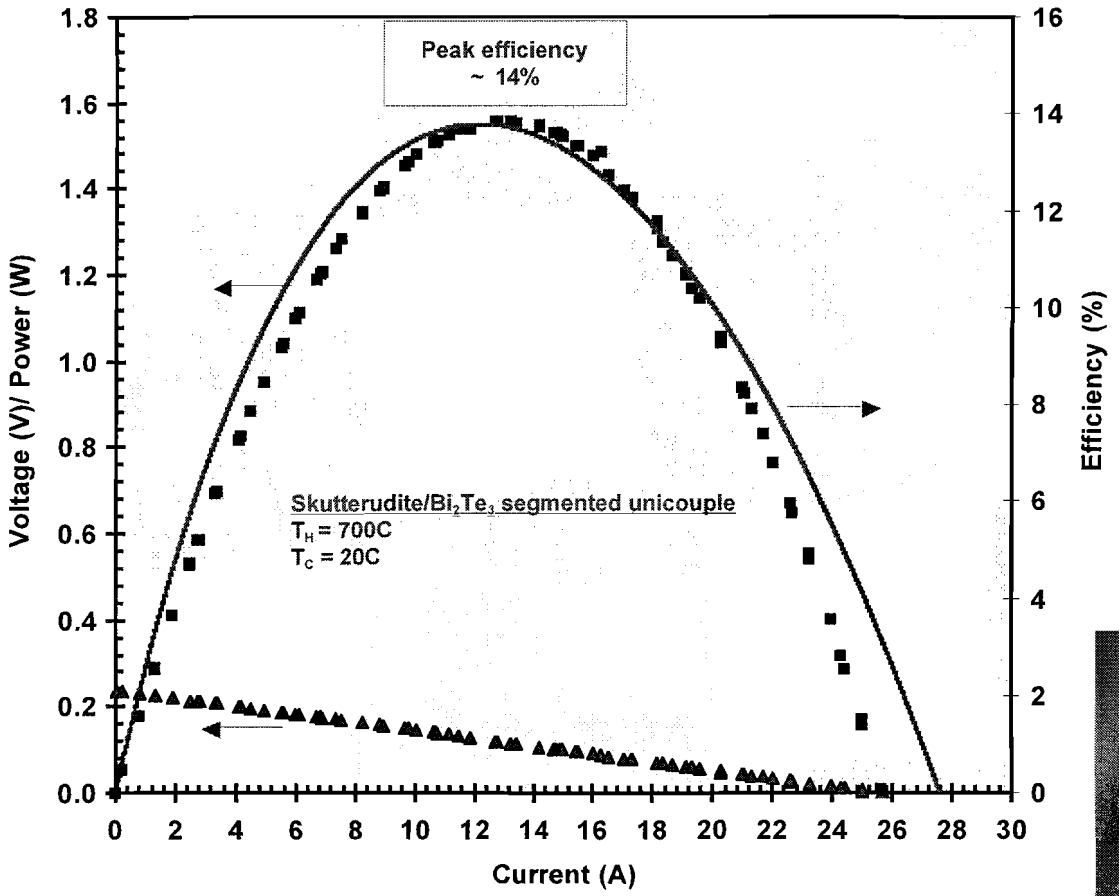


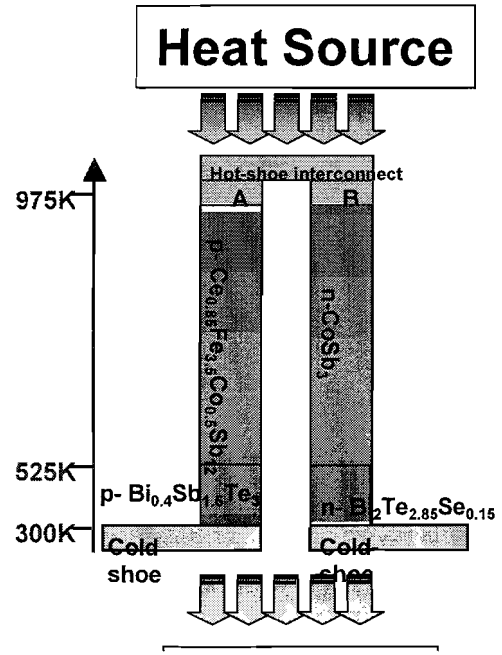
Skutterudite unicouples fabricated by diffusion bonding

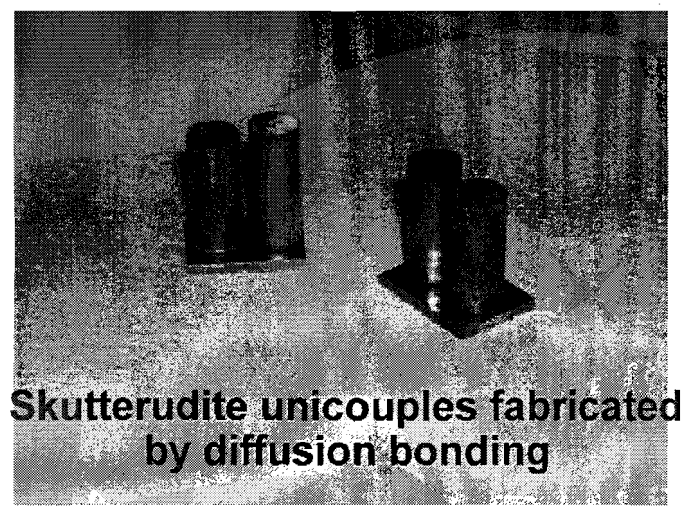
- **Skutterudite unicouples**
  - Developed diffusion bonding process and set-up for unicouple fabrication
  - Started to explore co-hot-pressing as an alternate fabrication method
- **Segmented unicouples**
  - Fabricated several metallized legs n- and p-segmented legs
  - Diffusion bonding of full unicouples in progress

Fabricated first generation skutterudite unicouples for screening and life testing in FY05

Thermal and electrical testing - Segmented unicouple







- Achieved ~ 14% efficiency for 975K-300K ΔT
  - For fully segmented unicouple
- Fully validate projected performance

Lifetime performance model development FY04-06

THERMOELECTRIC PROPERTIES (FY04)

Examples :
- SiGe: dopant precipitation
- Fine grained SiGe: grain growth
- TAGS : compositional change

Testing:
- Coupons

Impact:
- Change in efficiency, P output

Solution:
- Composition, doping control

MATERIALS SUBLIMATION (FY04)

Examples :
- SiGe: Si & Ge
- TAGS, PbTe: Te, Se, Ag, Sb
- Skutterudites : Sb

Testing:
- Coupons

Impact:
- A/l, porosity, contact resistance, mechanical failure

Solution:
- Encapsulation, coatings
- More refractory materials

THERMAL INSULATION (FY05-06)

Examples :
- Si in MFI

Testing:
- Unicouple

Impact:
- Shorting
- Contamination

Solution:
- Processing and engineering control

THERMO-MECHANICAL INTEGRITY (FY05-06)

Testing:
- Unicouple

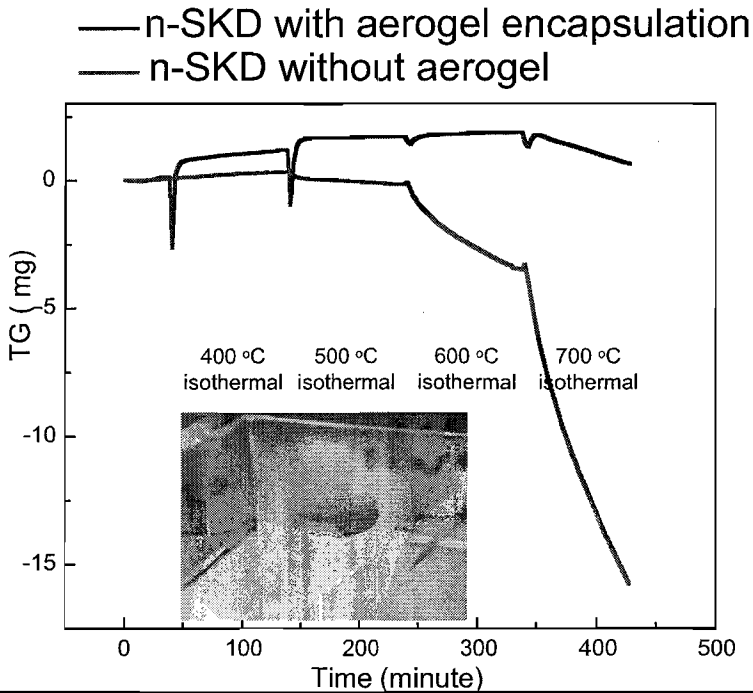
Impact:
- Contact resistance
- Mechanical failure

Solution:
- Device engineering/modeling

LIFETIME MODEL

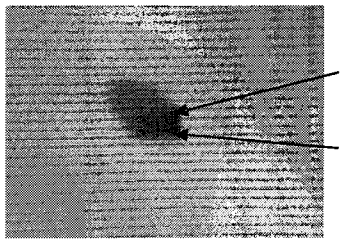
FUEL DECAY

Sublimation Suppression with Aerogel Encapsulation

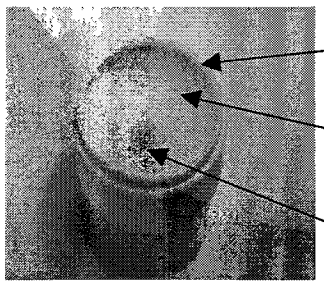


Sample	Temperature	Sublimation rate in vacuum (~ 2 _ 10^{-6} Torr)
n-SKD without aerogel	500 °C	5.31 _ 10^{-4} g/cm^2·hr
	600 °C	5.16 _ 10^{-3} g/cm^2·hr
	700 °C	2.15 _ 10^{-2} g/cm^2·hr
n-SKD with aerogel encapsulation	500 °C	-
	600 °C	-
	700 °C	2.70 _ 10^{-3} g/cm^2·hr

n-SKD cube (3 mm_3 mm_1.7mm) for the sublimation test



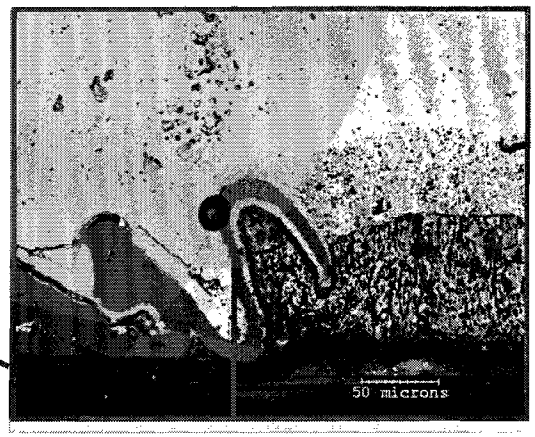
n-SKD cube encapsulated with aerogel (before sublimation test)



Bottom view of n-SKD cube encapsulated with aerogel (after sublimation test)

Ti coated-type tested in-gradient for 20 days

975K →

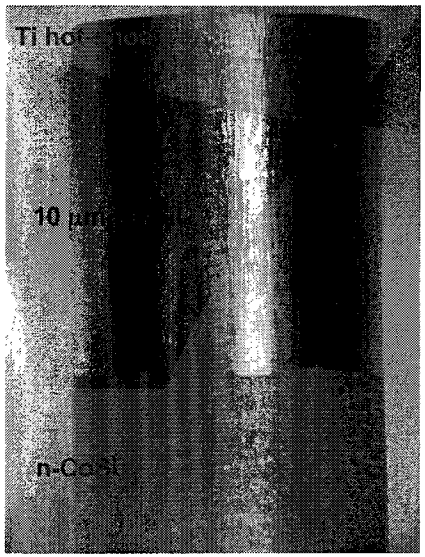


$CoSb_3$

$CoSb_2$

$CoSb$

Coating stops

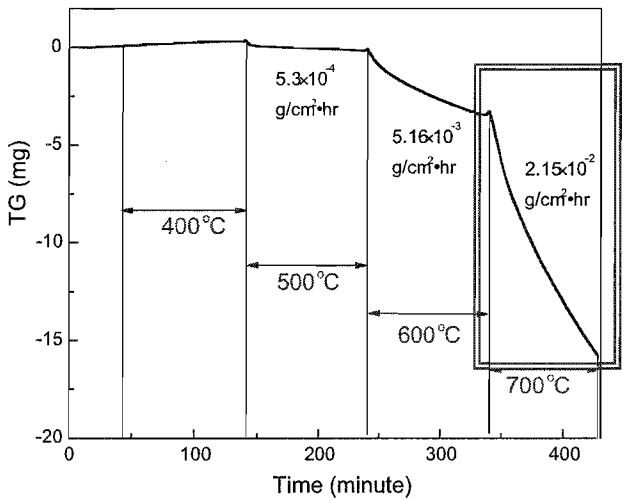


N-type Ti coated skutterudite leg

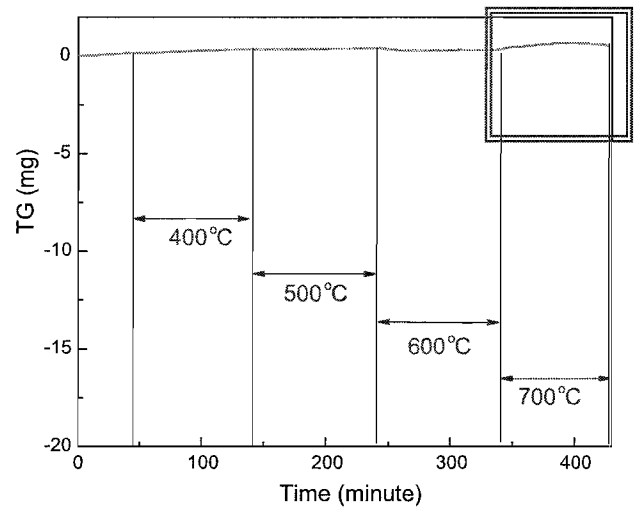
- No apparent degradation after 20 days
- Metal junction still intact
- Significant improvement over uncoated

FY 2004 Accomplishment: Demonstrated that Aerogel and Ti/Mo Coatings Significantly Reduce Sublimation at BOL

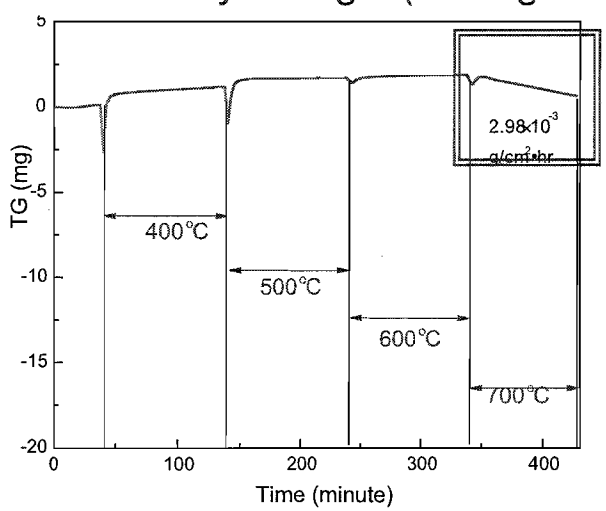
Uncoated



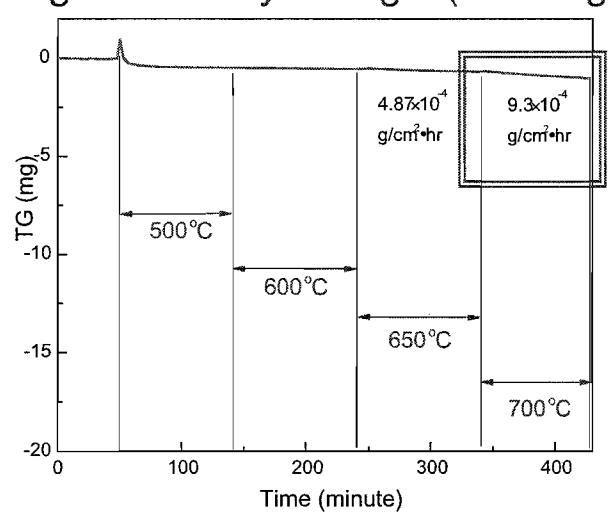
Titanium/Molybdenum Foil



Low Density Aerogel (50 mg/cm^3)



Higher Density Aerogel (100 mg/cm^3)



Sublimation rates from TGA measurement in dynamic vacuum

High temperature skutterudite sublimation rates at beginning of test

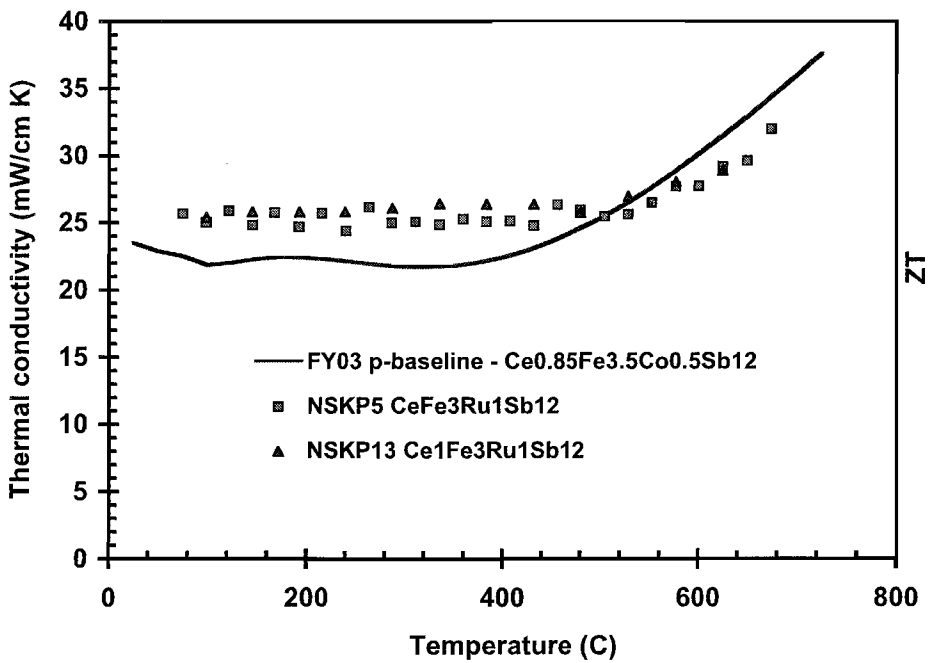
	Sublimation rate of n-SKD (ISN03)	Sublimation rate of p-SKD ($CeRu_4Sb_{12}$)
600 °C	3.88 _ 10^{-5} (g/cm^2·hr)	1.32 _ 10^{-4} (g/cm^2·hr)
700 °C	9.61 _ 10^{-4} (g/cm^2·hr)	7.91 _ 10^{-4} (g/cm^2·hr)
800 °C	3.92 _ 10^{-3} (g/cm^2·hr)	1.65 _ 10^{-3} (g/cm^2·hr)
900 °C	1.66 _ 10^{-2} (g/cm^2·hr)	1.05 _ 10^{-2} (g/cm^2·hr)

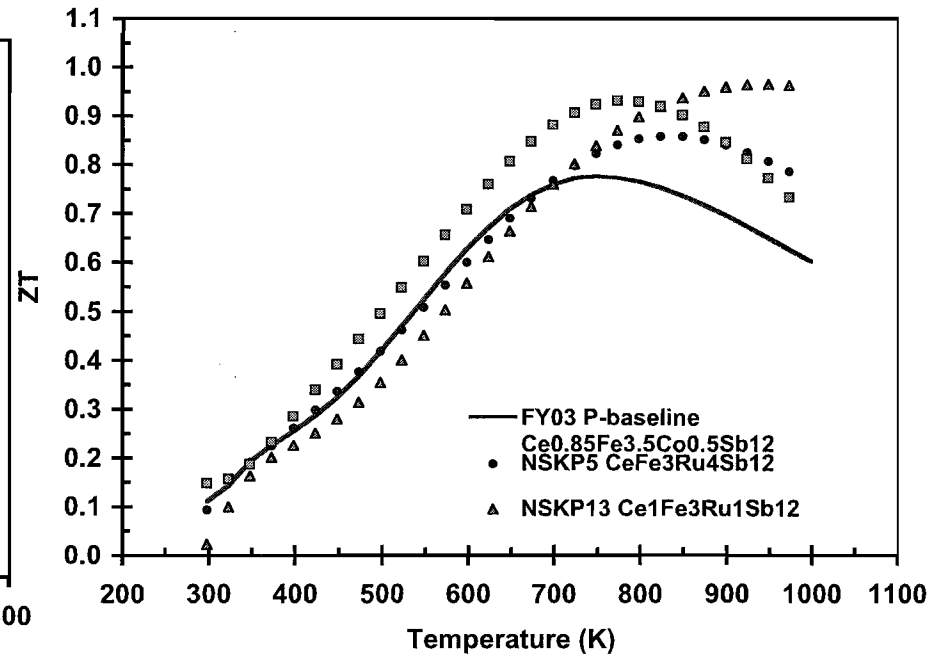
Skutterudite sublimation rates at beginning of test

	Sublimation rate of n-SKD	Sublimation rate of baseline p-SKD ($Ce_{0.85}Fe_{3.5}Co_{0.5}Sb_{12}$)	Sublimation rate of new p-SKD ($Ce_1Fe_3Ru_1Sb_{12}$)
500 °C	5.31 _ 10^{-4} (g/cm^2·hr)	7.40 _ 10^{-4} (g/cm^2·hr)	1.70 _ 10^{-4} (g/cm^2·hr)
600 °C	5.16 _ 10^{-3} (g/cm^2·hr)	6.78 _ 10^{-3} (g/cm^2·hr)	4.12 _ 10^{-4} (g/cm^2·hr)
700 °C	2.15 _ 10^{-2} (g/cm^2·hr)	2.80 _ 10^{-2} (g/cm^2·hr)	1.40 _ 10^{-3} (g/cm^2·hr)

Decreasing sublimation rates

Thermal conductivity and ZT versus temperature for n-type materials

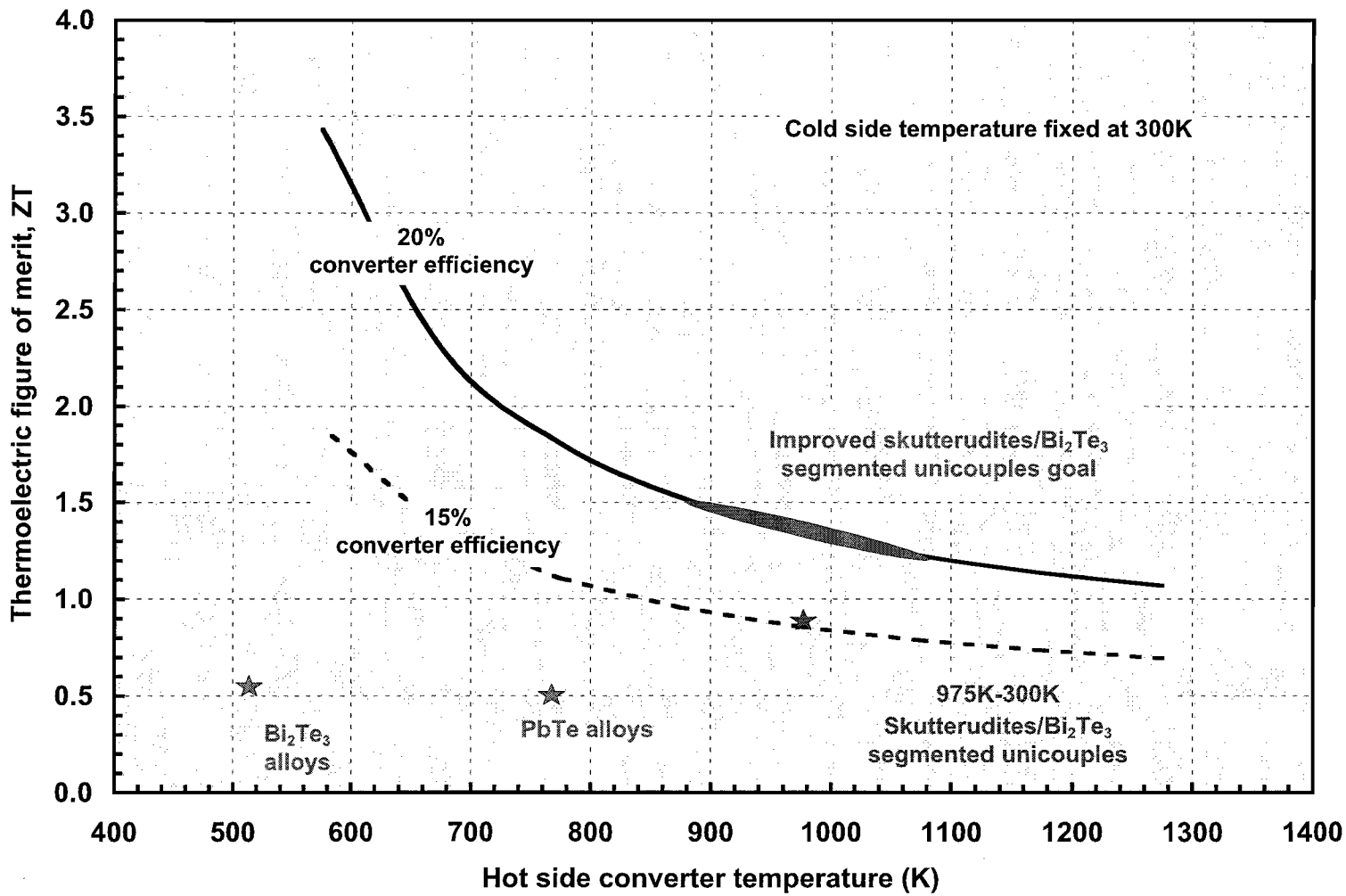




- Developed second generation p-type $CeFe_3Ru_1Sb_{12}$TE material that possesses improved ZT and higher temperature stability (1100K) than $Ce_{0.85}Fe_{3.5}Co_{0.5}Sb_{12}$ baseline material

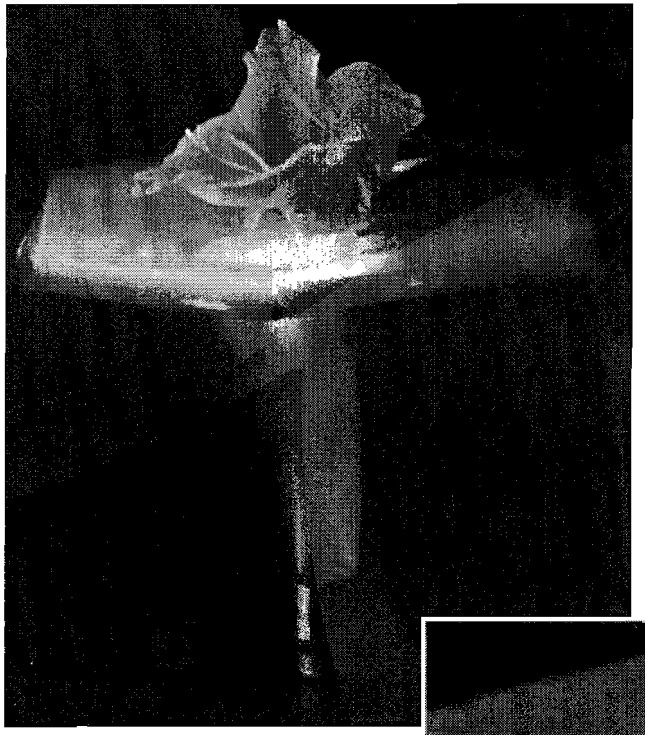
☞ $CeFe_3Ru_1Sb_{12}$TE material is now being used as the new p-type high temperature segment

Achieving 20% Converter efficiency



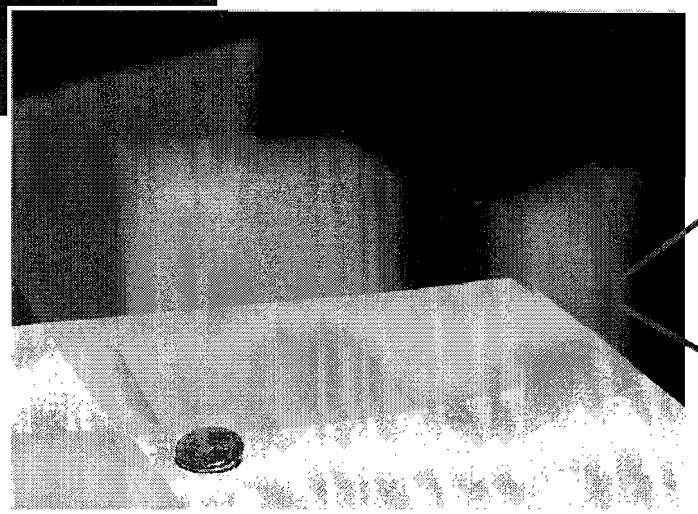
Back-up charts

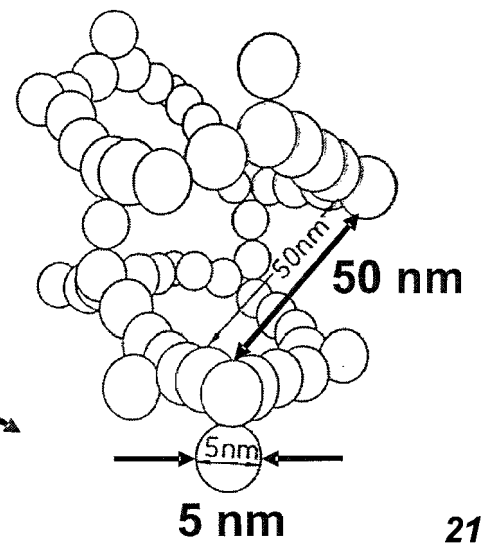
Aerogels



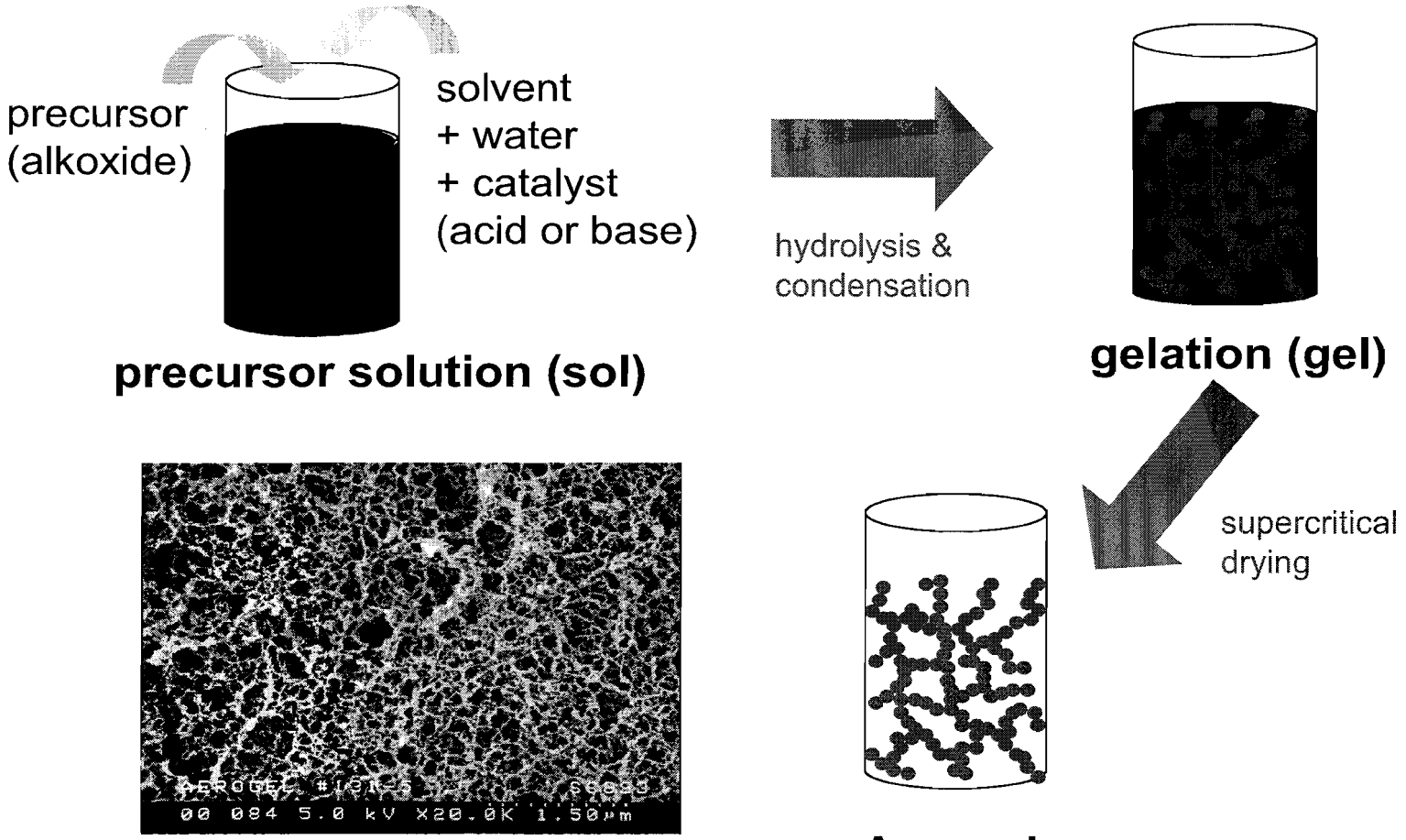
Lawrence Berkeley Lab

- Aerogels have a porous structure in which most volume is empty space (up to 99.9%)
- Lightest solid material in the world
- Excellent thermal insulator
- Excellent acoustic insulator
- Large surface area (500 ~ 1000 m^2/g)





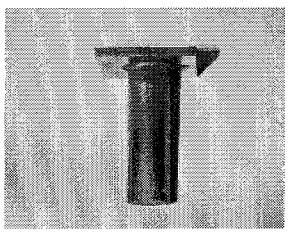
Processing of Aerogels



A SEM image of silica aerogel

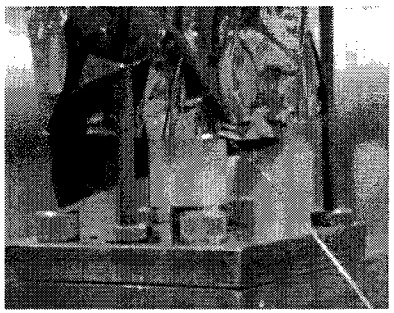
Sublimation Suppression November 2004

- Completed in-gradient experiment on an n-an p-type leg pressed at 535C and bonded to a Mo plate
  - A single p-leg (6 mm long) was bonded to a Mo plates
  - Tested in 700C-100C gradient for 7 days with a 4mm thick, machined, titania opacified/150 mg/cc aerogel donut
  - Post cross-sectional analysis showed that: both the leg and the aerogel indicated that there were no signs of significant sublimation (no noticeable depletion bands in the leg and no condensation inside the aerogel or on the inner wall of the ampoule

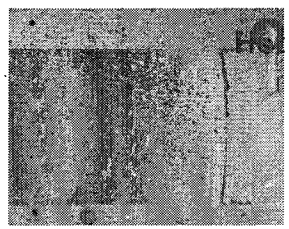


Single skutterudite leg bonded to a Mo hot-shoe

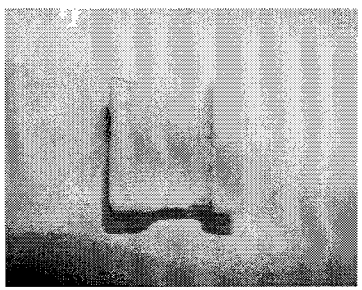
Bonded legs with machined aerogel donuts



In-gradient test set-up



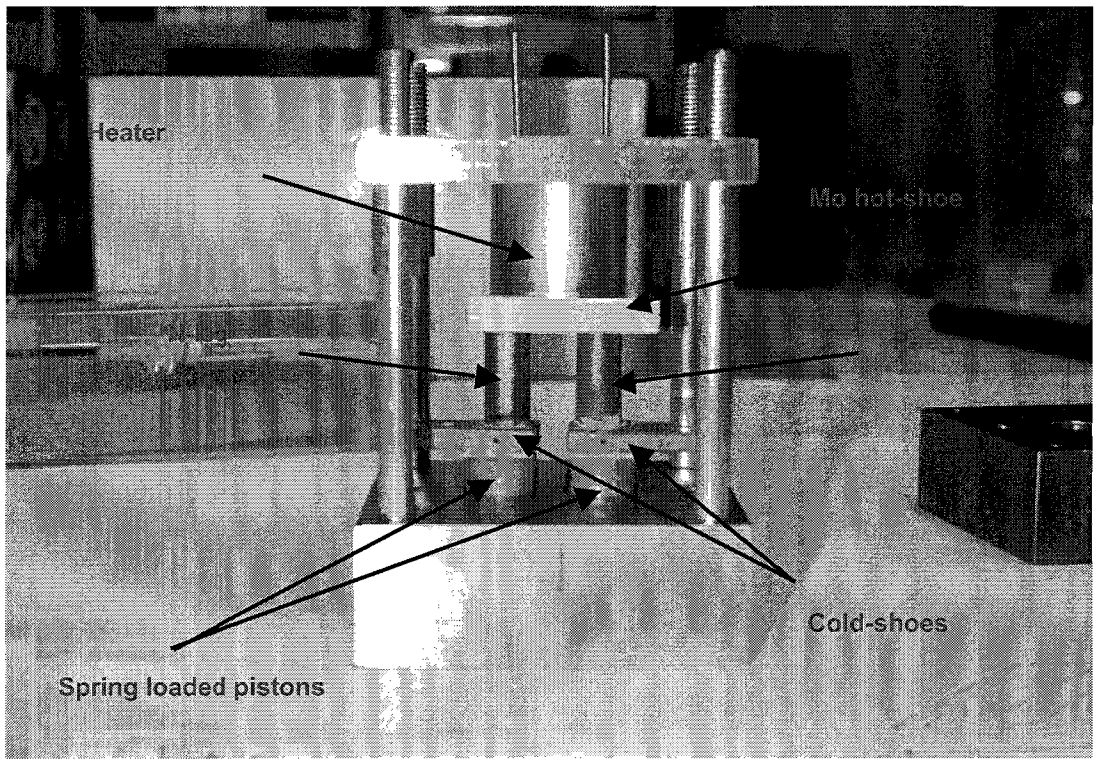
SEM picture of cross section near the Mo/leg interface after 7 days at 700C: no sublimation was observed



Photograph of aerogel donut cut in half after 7 days at 700C in contact with skutterudite leg: no sublimation deposit was observed

Spring loaded unicouple test fixture development

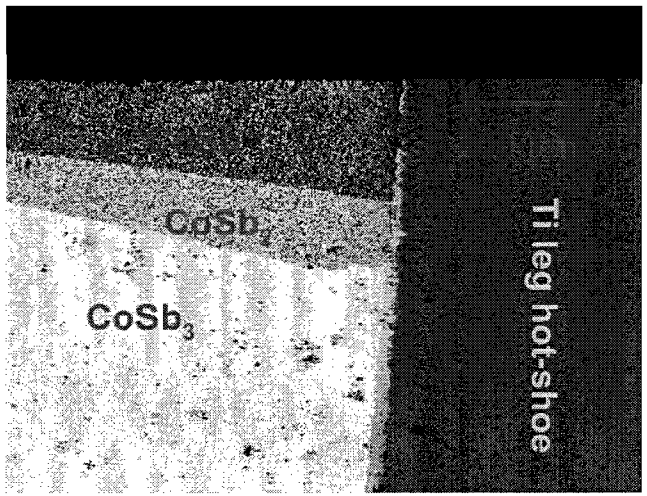
- **Fabricated 1st generation spring loaded test fixtures**



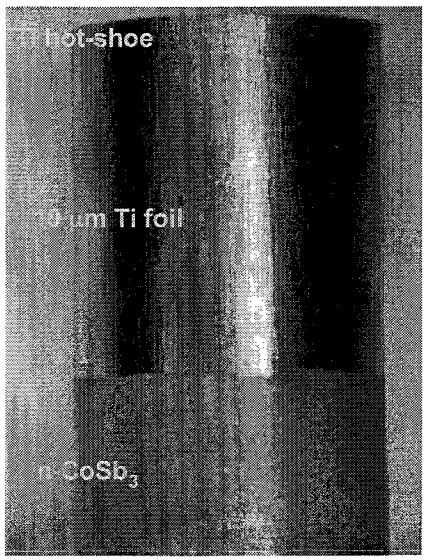
P- and n-skutterudite legs mounted in spring loaded test fixture

Sublimation control experiments

- Uncoated samples
  - Weight loss and temperature stability showed Sb sublimation in dynamic vacuum for T from ~ 875 to 975K for N-and p-type skutterudites
  - Decomposition into lower antimonide compounds
  - Appears to be diffusion limited



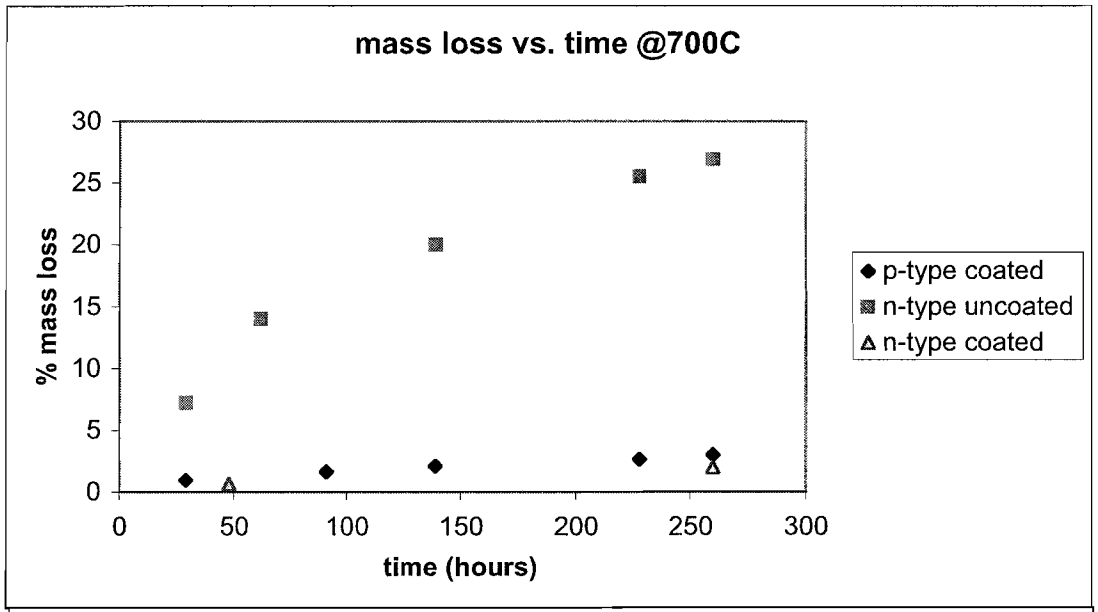
Photograph showing the decomposition of a $CoSb_3$ sample annealed at 875K for 3 months

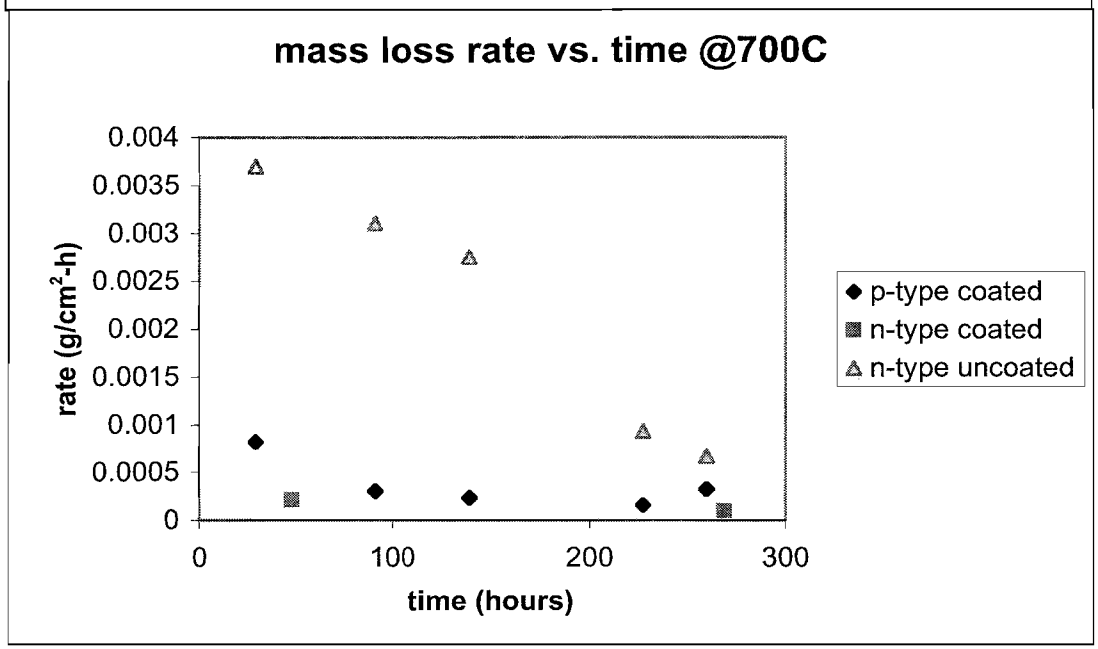


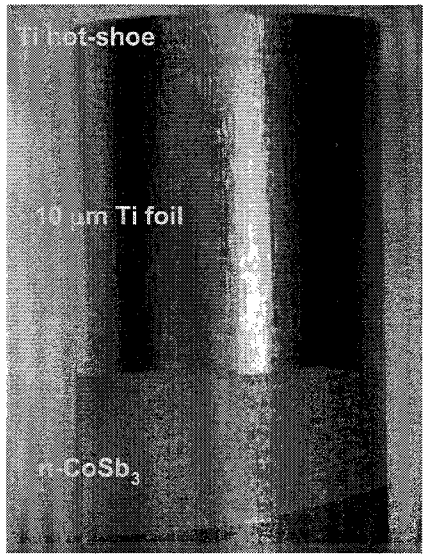
N-type Ti coated skutterudite leg

- Sb sublimation control studies
  - Use of cover gas significantly suppresses Sb sublimation
  - ~ 10 μm metallic film applied during hot-pressing
  - Performed in gradient Ti film-coated leg life tests
- Thermal/mechanical modeling performed to evaluate impact on unicouple performance

Weight loss experiments results



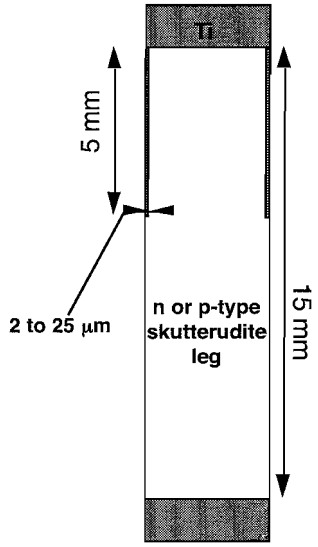


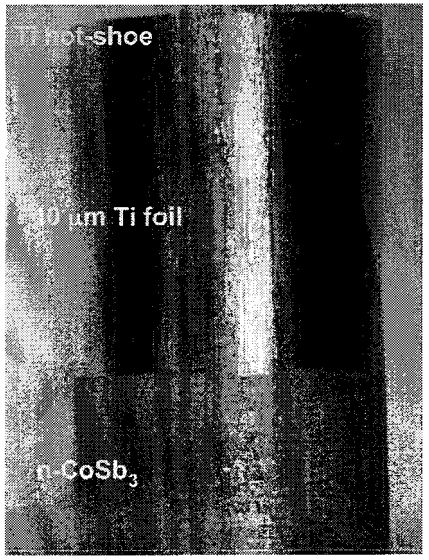


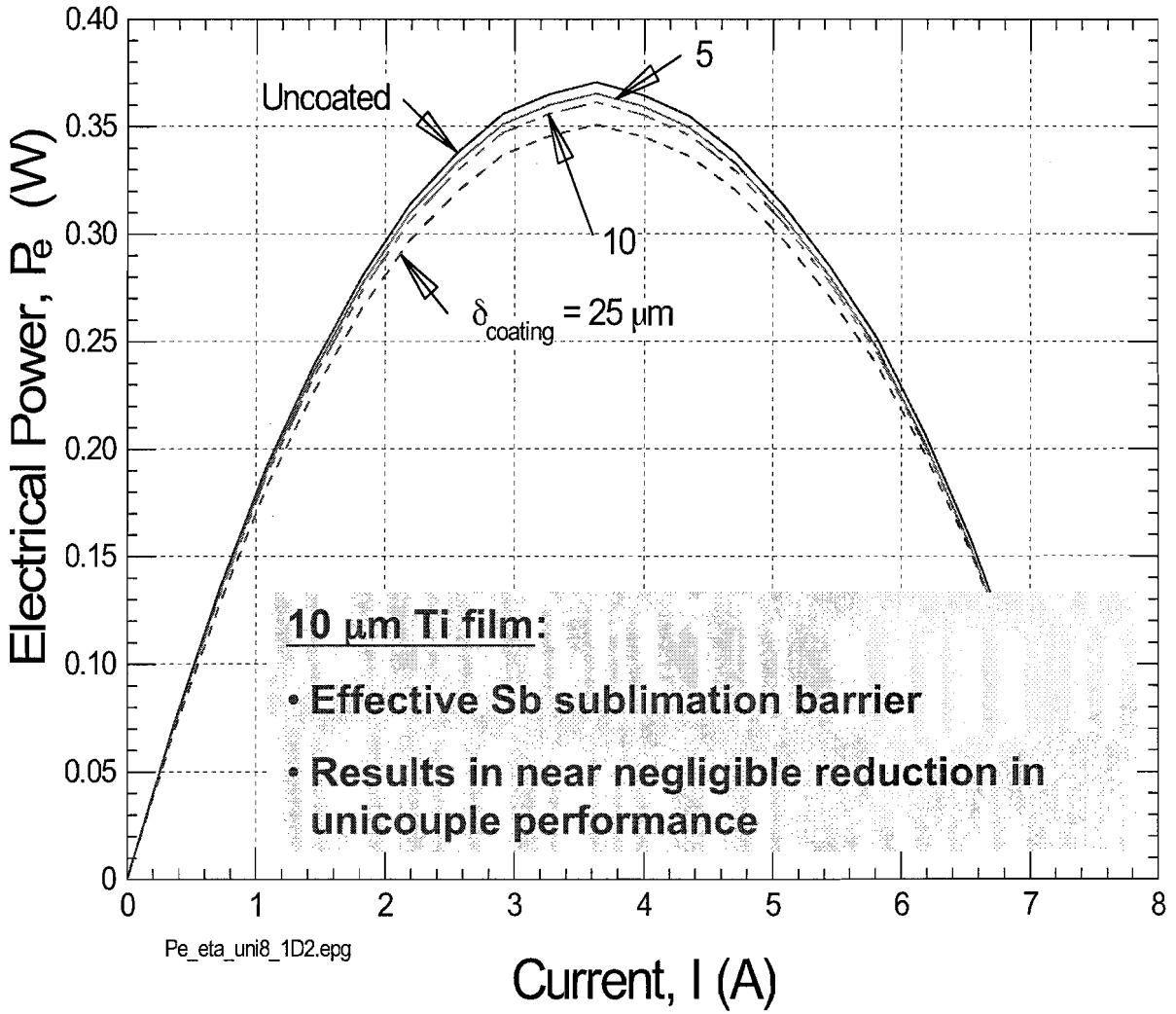
N-type Ti coated skutterudite leg

- Ti coating results in a significant mass loss reduction over uncoated

975K-375K Segmented TE Unicouple *performance modeling (UNM)*







10 μm Ti film:

- **Effective Sb sublimation barrier**
- **Results in near negligible reduction in unicouple performance**

Coating Material: Ti (length 5 mm and thickness = 0-25 μm)