Fabrication and Testing of Advanced Thermoelectric Unicouples for Power Generation

presented at the

21st International Conference on Thermoelectrics

Long Beach, CA
August 2002

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Program overview

- Engineering and testing
- Unicouple fabrication
- Modeling

- Materials Research
  - Develop high ZT materials
  - Materials synthesis
  - Improve thermal stability and compatibility

- High-efficiency prototype
Advanced Thermoelectrics DARPA & ONR effort at JPL

- Bulk materials
  - DARPA “Advanced Thermoelectrics” and ONR “Skutterudites”
    - Focus on TE materials for cooling and power generation up to 600-700°C
    - Identification, characterization and optimization of some new, promising materials
    - Skutterudites

![Diagram of Skutterudites structure]

La, Ce, Pr, Nd | Fe, Ru, Os, Co, Rh, Ir | P, As, Sb
Best ZT to date on new materials developed at JPL

- Combine best new material (Skutterudite) with the best SOA (BiTe)
State-of-the-art vs. advanced thermoelectric technology

- Projected performance is double SOA technology
Segmented legs: fabrication and testing

- **Segmented leg fabrication**
  - Uniaxial hot-pressing of powder stacked on top of each other
    - Temperature optimized → density close to theoretical value
    - In graphite dies and argon atmosphere
  - With metallic foil between segments
    - Selected to compensate for coefficient of thermal expansion mismatch
    - Diffusion barrier
    - Should react chemically with both materials to be bonded
    - Low electrical resistance bond (<10μΩcm²)
  - Metallic contacts at hot-side

- **Bond quality**
  - Electrical contact resistance measurement
  - Microprobe analysis
    - Diffusion
    - Chemical reaction and interface layer analysis

Segmented legs fabricated by uniaxial hot-pressing
In gradient, electrical contact resistance life tests on $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}/\text{CoSb}_3$ segmented leg (n-type)

Demonstrated low electrical resistance contacts between $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$ and $\text{CoSb}_3$ segments
In gradient voltage output and resistance measurements for $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}/\text{CoSb}_3$ segmented leg (n-type)

- Results validate thermoelectric properties of n-type $\text{CoSb}_3$ $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$ $\text{CoSb}_3$ segmented leg
- Confirms low electrical contact resistance between segments
In gradient electrical contact resistance life tests on $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$/CeFe$_4$Sb$_{12}$ segmented leg (p-type)

- Demonstrated low electrical resistance contacts between $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ and CeFe$_4$Sb$_{12}$ segments
Temperature stability tests have identified Sb sublimation as the predominant material dissociation mechanism in dynamic vacuum.

- Cover gas suppresses Sb sublimation
Sol-gel silica coatings

- To prevent sublimation in vacuum; use coating similar to SiGe technology
- Sol-gel; dip coating SiO$_2$ method
- 1 coat: <500 nm
- Non-conductive and thermally insulating
- Stable < 900°C
- Improve coating quality by minimizing TE porosity
Sol-gel coatings suppress Sb sublimation

- Sample heated to 600°C/dynamic vacuum/72 hours
- Single coat provides good protection
Unicouple testing

Alignment pins

W Heater

p n

cold sink
Primary objective: demonstrate 15% efficiency to match predicted performance

Consistently achieve > 10% efficiency

Hot-side, contact resistance must be reduced

Achieved by optimizing braze material and method of brazing
Potential applications

- Thermal to Electric Power Generation
  - Integration with any heat source
    - Combustors
    - Catalytic reactors
    - Radioisotope heat source

- Waste heat recovery
  - Automobile exhaust
    - Supplement or replace electrical power generator with electrical power generated from engine waste heat
    - $T \sim 600$ to 700$^\circ$C available at the catalytic converter
    - $\sim 1$ kW power generator
    - Cost is critical
  - Power plants
  - Geothermal energy
  - Jet engines

- Solid State Advantage
  - No moving parts
  - No maintenance
  - Long life
Summary

- New segmented thermoelectric unicouples under development
  - Operating between 300 and ~ 1000K
  - Predicted efficiency up to 15%

- Unicouple fabrication and testing
  - Several segmented and non-segmented unicouples built for thermal and electrical testing
  - 10% thermal to electrical efficiency routinely demonstrated
  - Several engineering and processing challenges remain

Acknowledgement

- NASA, DARPA and ONR