Thermoelectric Mechanical Reliability

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Overview

Timeline

- Project start: October 2006
- CRADA start: March 2009
- Project end date: March 2013 (1-yr extension pending)
- Percent complete: 85%

Budget

- Total project funding

	FY09	FY10	FY11	FY12
DOE	$300K	$300K	$300K	$375
Marlow Ind.	$75K	$150K	$150K	$150

Barriers

- Barriers addressed
  - 2/3 chemical energy in automotive fuel is rejected to atmosphere as waste heat
  - Thermomechanical stresses must be managed and TE material strength improved to fully exploit TE devices
  - TE materials are inherently brittle and susceptible to thermal-induced fracture
- Targets*
  - 5000h life or 10 yr or 150k mile lifetime
  - Brittle bulk materials must survive thermal and mechanical stresses for life

Partners

- Marlow Industries (CRADA)
- General Motors (indirectly)

* "A Science-Based Approach to Development of Thermoelectric Materials for Transportation Applications, Office of FreedomCAR and Vehicle Technologies, August 8, 2007.

Objectives

- Measure needed thermomechanical and thermophysical properties of candidate thermoelectric materials (TEMats) considered for waste heat recovery and cooling applications in vehicular applications.
- Combine measured data with established probabilistic reliability and design models to optimally design automotive and heavy vehicle thermoelectric devices (TEDs) for heat recovery and cooling.
- Coordinate international round-robin testing and measurement of transport properties with the intent to improve their consistency and validity.

Milestones

- **FY11:**
  - **Generate thermoelastic and mechanical property database as a function of temperature on Marlow's candidate p- and n-type TEMats as they are developed**
  - **Provide mechanical characterization of other material constituents used in the TEDs.**
- **FY12:**
  - **Measure mechanical, thermoelastic, and thermoelectric properties of Marlow-fabricated TEMats to enable operation up to 500°C.**
  - **Complete report of international round-robin test results on Marlow bismuth telluride to IEA-AMT and initiate a new high temperature thermoelectric round-robin measurement on PbTe or skutterudite ranging between 20-500°C.**

Technical Approach

- Measure Young's Modulus, Poisson's ratio, CTE, thermal conductivity, heat capacity, and strength as a function of temperature of candidate Marlow (and General Motors) TEMats.
- Perform fractography on strength specimens, identify failure initiation sites and strength-limiting flaw types, and recommend processing recommendations that will improve strength.
- Use probabilistic design and reliability methods with candidate and prototype TEDs.
- Provide mechanical evaluation of the other material constituents used in Marlow's TEDs.
- Coordinate international round-robin testing and measurement of transport properties with the intent to improve their consistency and validity.

Technical Accomplishments – 1 of 11

- Strength tested hundreds of bend bars fabricated by Marlow as part of ORNL-Marlow CRADA
- Evaluated stresses in metallization used with TEDs as part of ORNL-Marlow CRADA
- Neutron diffraction explored as a means to estimate residual stresses in thermoelectric legs in devices
- Coordinated international round-robin testing and measurement of transport properties.

Why is Mechanical Strength Important to TEMats?

$$R_{Therm} = \frac{S_{Tens}(1-\nu)\kappa}{CTE \bullet E}$$

Kingery, J. Am. Cer. Soc., 38:3-15 (1955).

R_{Therm} = Thermal resistance parameter (the larger the better)
S_{Tens} = Tensile stress or strength
ν = Poisson's ratio
κ = Thermal conductivity
CTE = Coefficient of thermal expansion
E = Elastic modulus

Griffith Criterion

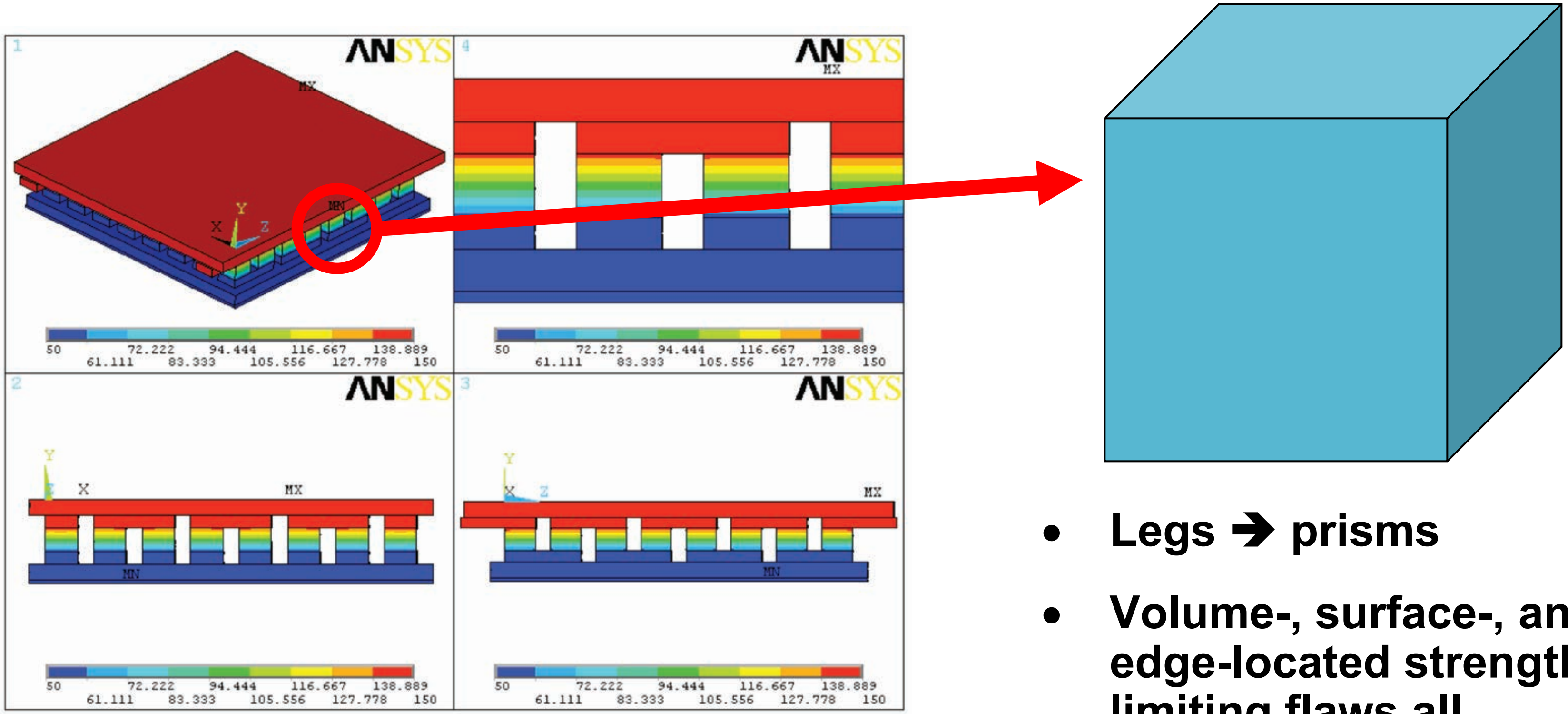
$$S_{Tens} = \frac{K_{Ic}}{Y\sqrt{c}}$$

KIc = Fracture toughness
Y = Crack shape factor
c = Griffith flaw size

Tensile Strength << Compressive Strength
Manage tensile stress for conservative design

Must seek to minimize c!

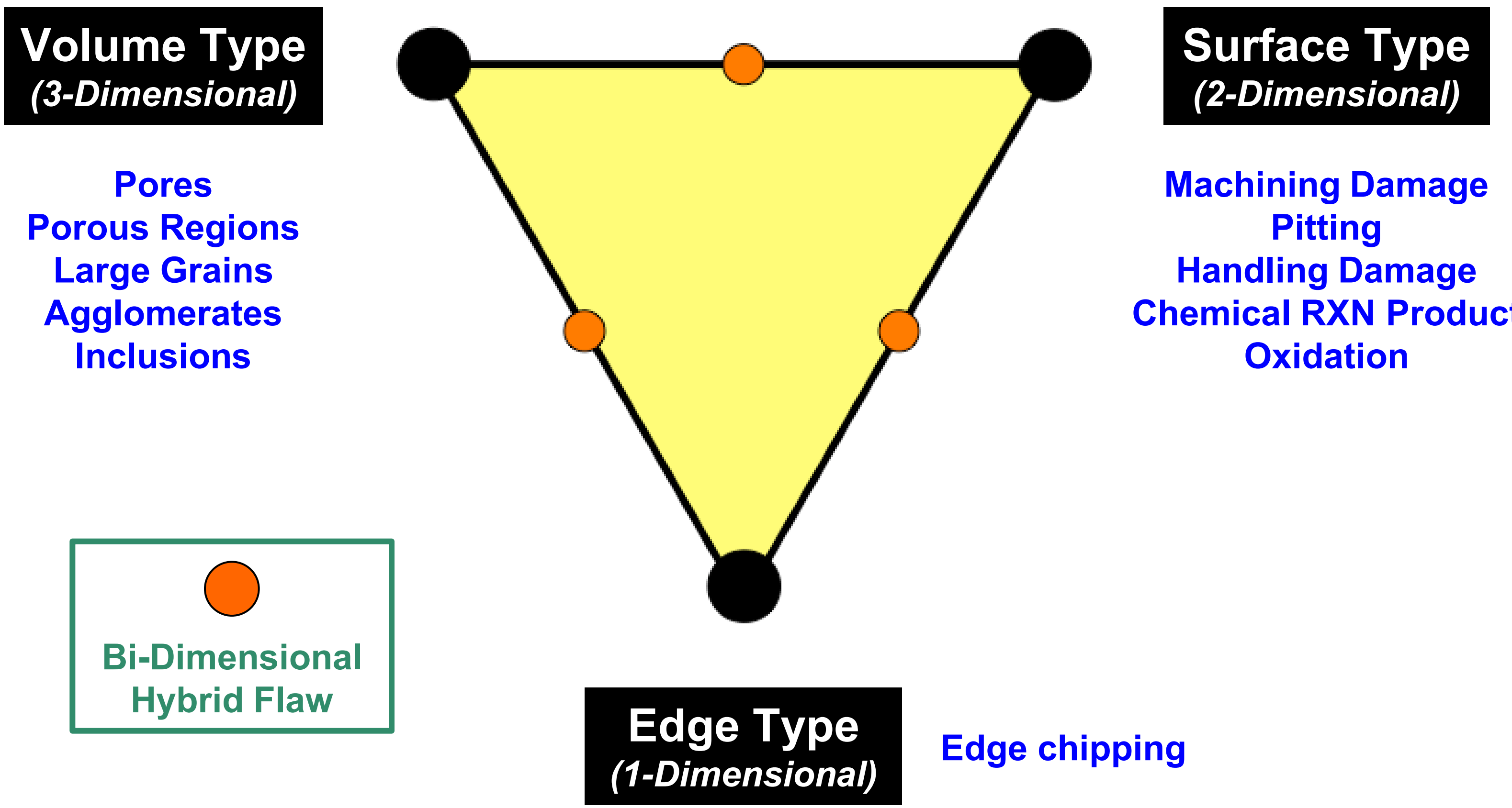
TE Legs and Potentially Active Flaws:



- **Legs ➔ prisms**
- **Volume-, surface-, and edge-located strength-limiting flaws all possibly active**
- **e.g., 3 x 3 x 3 mm**

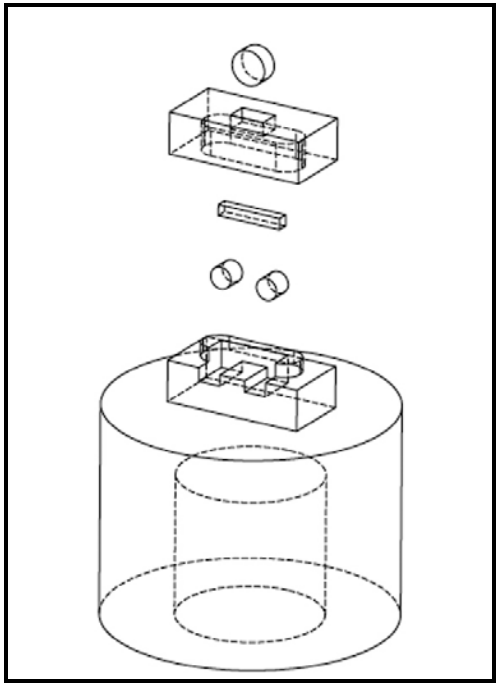
Technical Accomplishments – 4 of 11

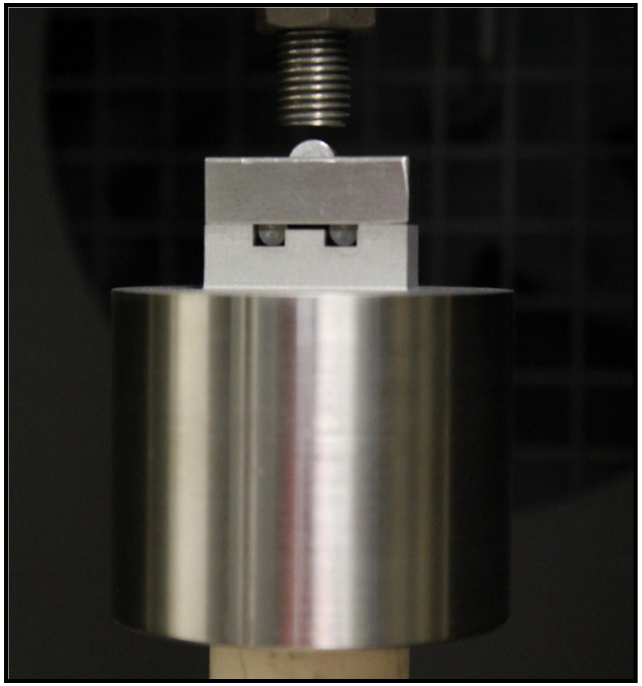
Strength-limiting flaw classification for brittle materials; the potential existence of all are in unchamfered TE Legs

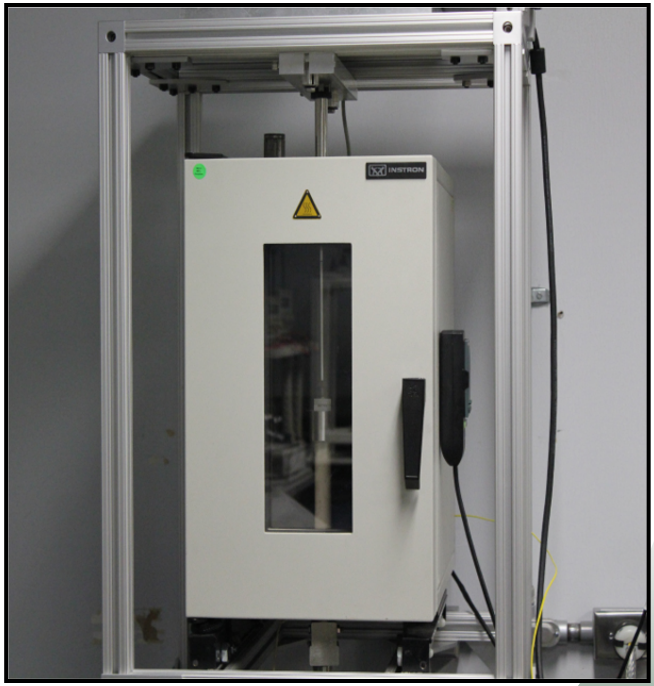


Technical Accomplishments – 5 of 11

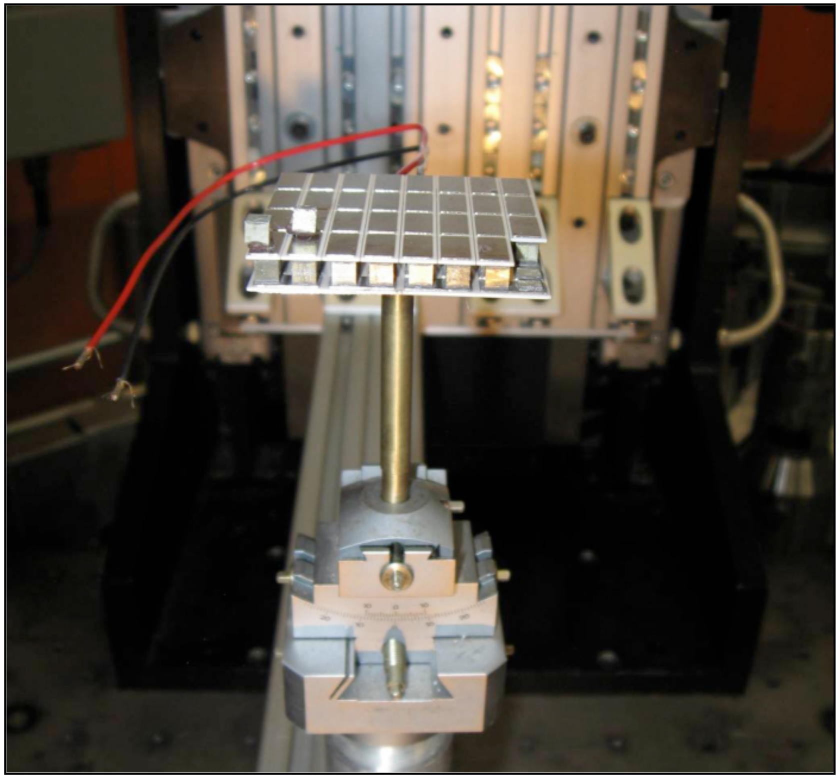
- There was a need in FY11 to test hundreds of specimens at T < 300°C
- New fixturing was designed and fabricated to facilitate rapid and valid flexure testing of TEMat bend bars

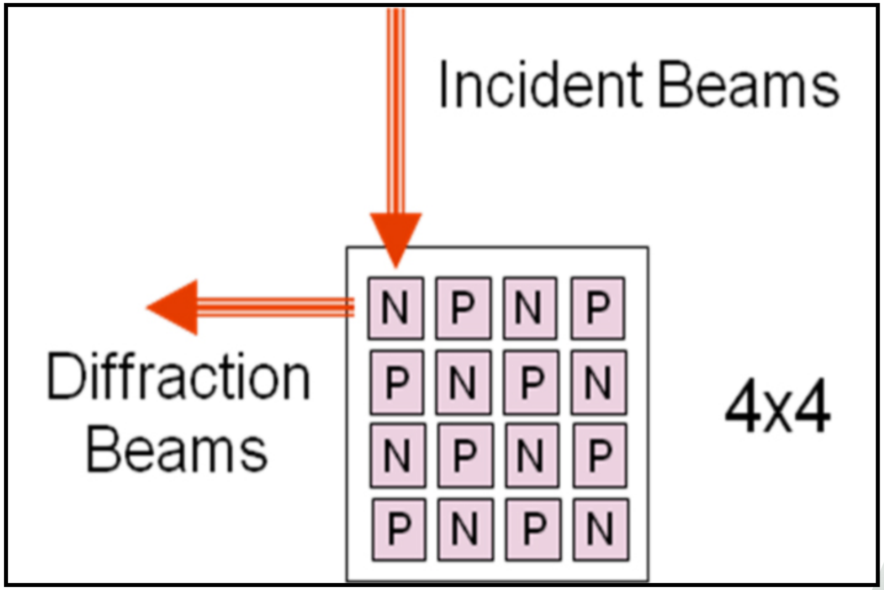






Neutron Diffraction Used to Measure Residual Stresses in Thermoelectric Devices





IEA-AMT Annex VIII Support and Participants

- **IEA-AMT Thermoelectric Annex**
  - Annex lead: Oak Ridge National Laboratory (H. Wang)
  - USA: Clemson (T. Tritt, S. Zhu); Marlow (J. Sharp); Corning (A. Mayolet, C. Smith, J. Senawiratne) and ZT-Plus (F. Harris)
  - China: SICCAS (SQ Bai, L. Chen)
  - Canada: Natural Resource Canada (J. Lo); University of Waterloo (Holger Kleinke); University of Quebec at Chicoutimi (Laszlo Kiss)
  - Germany: Fraunhofer IPM (H. Böttner, J. König)
  - International Observer: Japan: AIST (R. Funahashi)
  - International Observer: Korea: KERI (H. W. Lee)
- **ORNL Support: HTML (E. Lara-Curzio), W.D. Porter, and W. Cai**

Technical Accomplishments – 8 of 11

Marlow Materials Selected for Transport Properties Round-Robin Tests

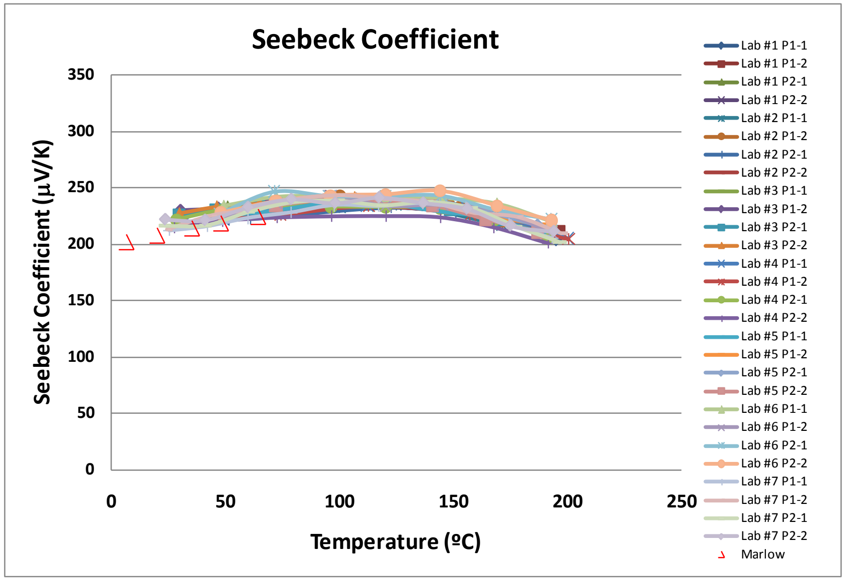
- Materials: $Bi_2Te_{3.005}$ (n-type) $Bi_{0.5}Sb_{1.5}Te_3$ (p-type)
- Four-sample Sets
  - Thermal diffusivity: 12.7 mm diameter disk
  - Specific heat: 4 mm diameter disk
  - Seebeck coefficient and electrical resistivity: 2 x 2 x 15 mm^3 bar, 3 x 3 x 12 m^3 bar
- Temperature range: 20-200ºC
- Round-robin plans:
  - Use best practice in each lab
  - Focusing on one specific material
  - Develop test procedures
  - Round-robin 1 focus: n-type and p-type materials
  - Round-robin 2 focus: p-type materials

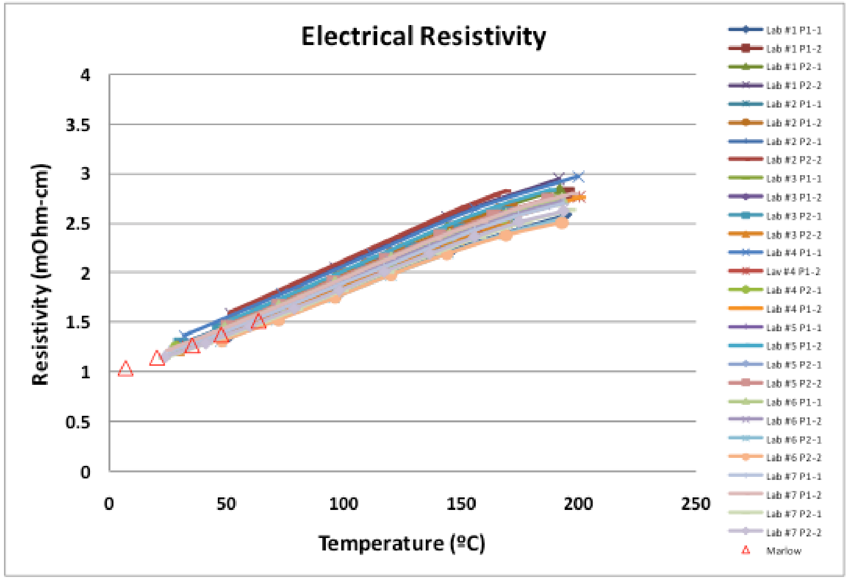
Technical Accomplishments – 9 of 11

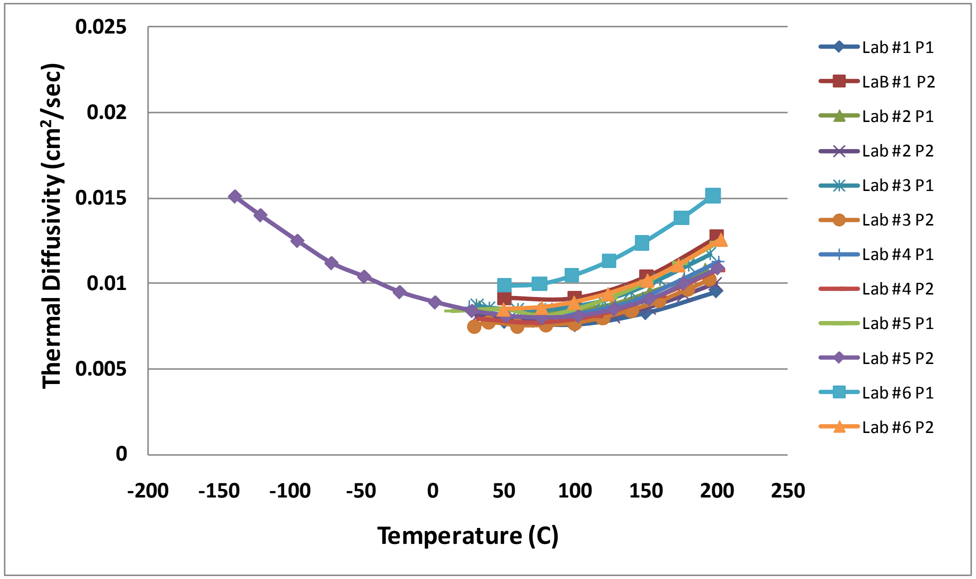
Round-Robin #2 Started in October 2010

- Procedures for DSC prepared by ORNL
- Two sets of p-type samples
  - Set #1: ORNL -> Clemson-> Corning -> ZT-Plus -> Germany -> China -> Canada
  - Set #2: China -> (Japan) -> Germany -> ORNL -> Clemson-> Corning -> ZT-Plus -> Canada
- Completed in July 2011
- Report to IEA-AMT: October 2011

Technical Accomplishments – 10 of 11







Technical Accomplishments – 11 of 11

Round-robin observations so far:

- IEA-AMT is addressing the important issue of measurement and standardization of thermoelectric properties
- Significant measurement issues were observed especially in specific heat and electrical resistivity
- P-type material was selected for 2nd round robin test
- Good agreements in Seebeck coefficient and electrical resistivity
- Thermal diffusivity in good agreement expect for one test (data analysis)
- Specific heat remains an issue for reliable ZT

Future Work

- **Continue to collaborate with Marlow Industries, a manufacturer of high-temperature-capable TEMats and TEDs, to contribute to the reliability improvement of their candidate TEMats**

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- **Mechanically evaluate constituents (e.g., interconnects) in TEDs and TEDs (themselves)**

- **Support other mechanical reliability issues associated with the TEDs, for example, combating potential residual stresses associated with metallization**

Summary

- **Property evaluations**
  - Flexure strength evaluated of refined Marlow TEMats.
  - Residual stresses estimated in metallization layers in Marlow TEDs.
  - Neutron diffraction explored as means to estimate residual stresses.

- **Property measurement consistency**
  - International round-robins examining property measurements of relevant parameters.
  - Heat capacity measurement a potential issue.

- **Testing in FY12**
  - Mechanical property evaluation of next Marlow TEMats.
  - TED thermal measurement estimations underway.
  - Mechanical characterization of other material constituents in TEDs. Shear testing of interconnects.