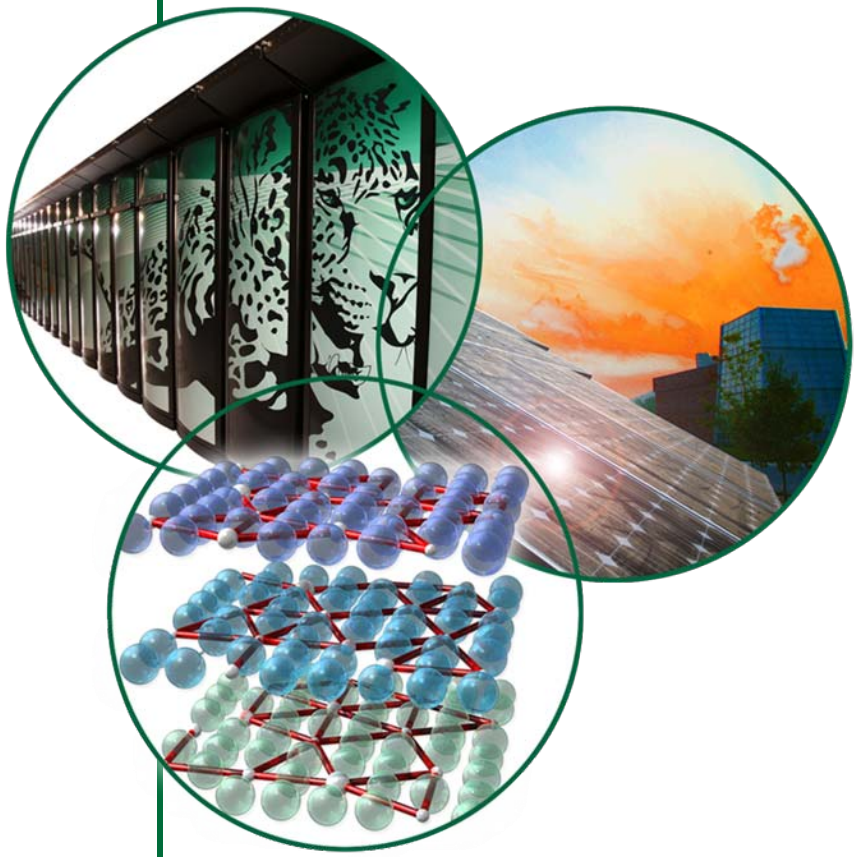
Project ID: LM030, Wang

High-Temperature Thermoelectric Materials Characterization for Automotive Waste Heat Recovery: Success Stories from the High Temperature Materials Laboratory (HTML) User Program



DOE 2010 Vehicle Technologies Annual Merit Review and Peer Evaluation Meeting

Dr. Hsin Wang and Dr. Miaofang Chi
Dr. Ralph Dinwiddie
HTML User Program
Materials Science and Technology Division
Oak Ridge National Laboratory

Washington, DC
June 9, 2010

Sponsored by
U.S. Department of Energy, Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Vehicle Technologies

U.S. DEPARTMENT OF ENERGY

This presentation does not contain any proprietary, confidential, or otherwise restricted information.

OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

The HTML User Program: Background

- The HTML is a National User Facility that supports the missions of DOE, EERE and the Vehicle Technologies Program in particular, by working with industry, universities, and other national laboratories to develop energy-efficient technologies that will enable the U.S. to use less petroleum. The HTML is organized into six user centers, which are clusters of highly skilled staff and sophisticated, often one-of-a-kind instruments for materials characterization.
- Access to the HTML User Program is provided through the HTML User Program proposal process. Research proposals are reviewed by a committee and approved based on scientific merit, relevance of the proposed research to the mission of DOE's Vehicle Technologies Program, and feasibility. Projects have a well-defined scope, and research is completed within 24 months and normally involves one or more user visits to the HTML.
- Both nonproprietary and proprietary research is conducted within the HTML User Program. There are generally no charges for nonproprietary research projects, and users conducting nonproprietary research must agree to submit research results for publication in the open, refereed literature. A nonproprietary project is complete when the results are published in the open literature and/or presented at a professional conference. For proprietary research, the user owns the research data, and all costs at the HTML are paid by the user based on DOE guidelines for ORNL costs.

The HTML User Program – FY2009 Activity

During FY2009, the HTML User Program collaborated with 11 companies, 14 universities, and 3 national laboratories on 41 user projects addressing critical technical barriers to achieving the goals of DOE's Vehicle Technologies Program. There were 107 researchers who visited the HTML for a total of 562 days to conduct experiments.

The HTML User Program FY2009 budget was $5,066,946 and was allocated as follows:

- Capital equipment: $514,025
- Operations: $4,552,921

Users cost-share their HTML user projects through:

1) direct involvement with HTML staff members during the development of the user project;
2) funding their travel to the HTML to perform research;
3) cost of materials provided by the user or the research performed prior to the user project;
4) collaboration with HTML staff members to analyze the data and publish the results.

The HTML also supports the education and preparation of a new generation of scientists and engineers.

During FY2009, students and professors from 14 universities participated in the HTML User Program. Four of those students earned their Ph.D. degree and three earned their M.S. degree based in part on research they conducted through the HTML User Program.

Relevance to the VT Program

- The Vehicle Technologies Program funds the operation of the HTML User Program to maintain world-class expertise and instrumentation capabilities for materials characterization to work with industry, universities and national laboratories toward the goals of the Vehicle Technologies Program. The HTML User Program capabilities also support the activities of the Vehicle Technologies Program's subprograms in Lightweight Materials, Propulsion Materials, Energy Storage, and Thermoelectric Conversion at the Oak Ridge National Laboratory.

- This poster presentation highlights ***one*** of the 41 user projects managed by the HTML User Program during FY2009. The user project in this poster presentation addresses the need for developing techniques for measuring key thermophysical and transport properties of thermoelectric materials. Material characterization also helps to achieve the goal of technology deployment by providing critical parameters for scale-up of thermoelectric modules. Materials with high ZT value are essential to achieving the goal of improving fuel efficiency by 10% or more through recovery of energy from engine exhaust and/or the engine cooling system.

General Motors R&D Center User Project:
"Thermoelectric properties of clathrates through a systematic cross-substitution of framework elements"

Timeline

- Start date: 10/1/08
- End date: 9/30/09
- % complete: 100%

Barriers

- Scale-up to a practical thermoelectric device
- High figure of merit (ZT)
- Lack of standard for property measurements

Budget

- Included in the user center allocations from the annual budget of the HTML User Program; users cost-share as noted on slide #3.

Collaborators

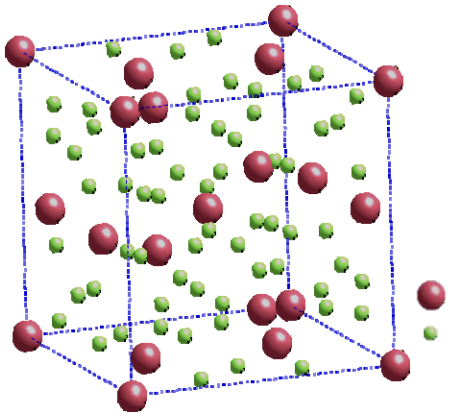
- **Users:** General Motors R&D Center Xun Shi, Jihui Yang, James Salvador
- **HTML Staff:** Hsin Wang, Miaofang Chi
- **GM Partners** at NIST and Shanghai Institute of Ceramics

General Motors R&D Center User Project

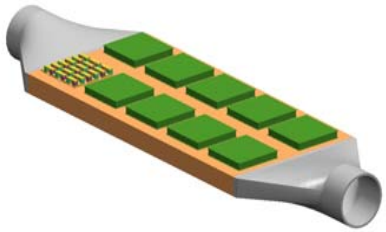
Research Problem: To determine the high-temperature transport properties of clathrates including thermal conductivity, electrical resistivity, and Seebeck coefficient; study atomic-scale structure.

Technical Approach: Utilize the unique capabilities of the HTML User Program for the evaluation and characterization of thermoelectric materials to obtain key transport properties and information about their atomic structure.

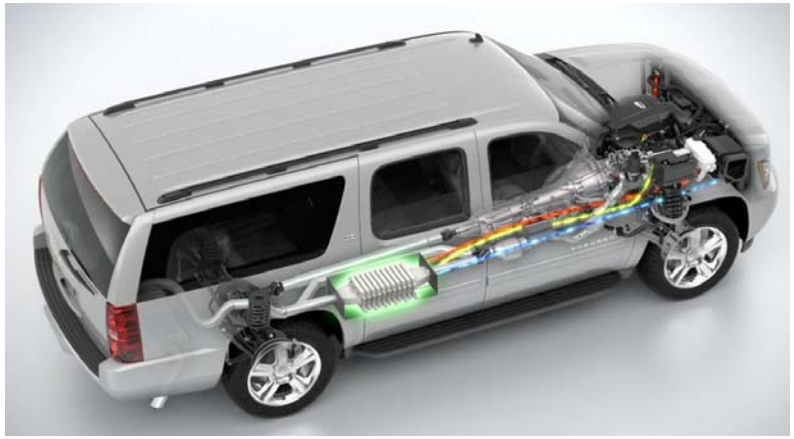
Materials



Devices



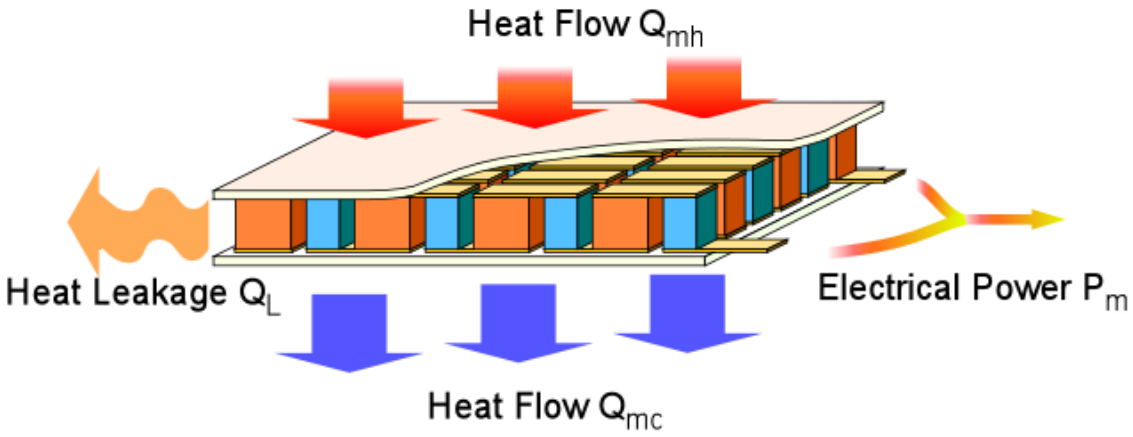
Vehicle Application



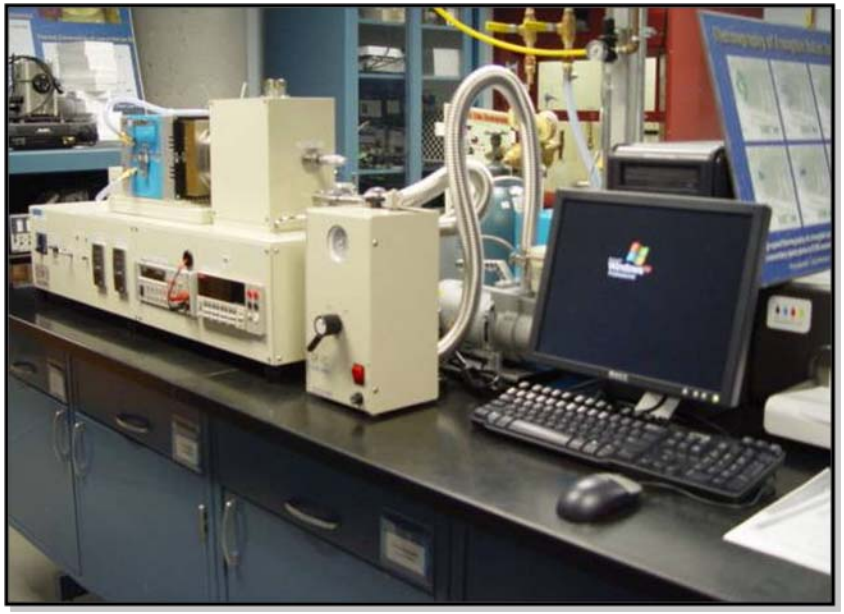
Schematic and photo courtesy of General Motors R&D Center.

General Motors R&D Center User Project: Milestones

- Characterize and obtain accurate thermophysical and transport properties for thermoelectric materials during the user visit
- Conduct atomic-scale imaging using STEM
- Jointly report the results of the user project via papers and presentations



General Motors R&D Center User Project



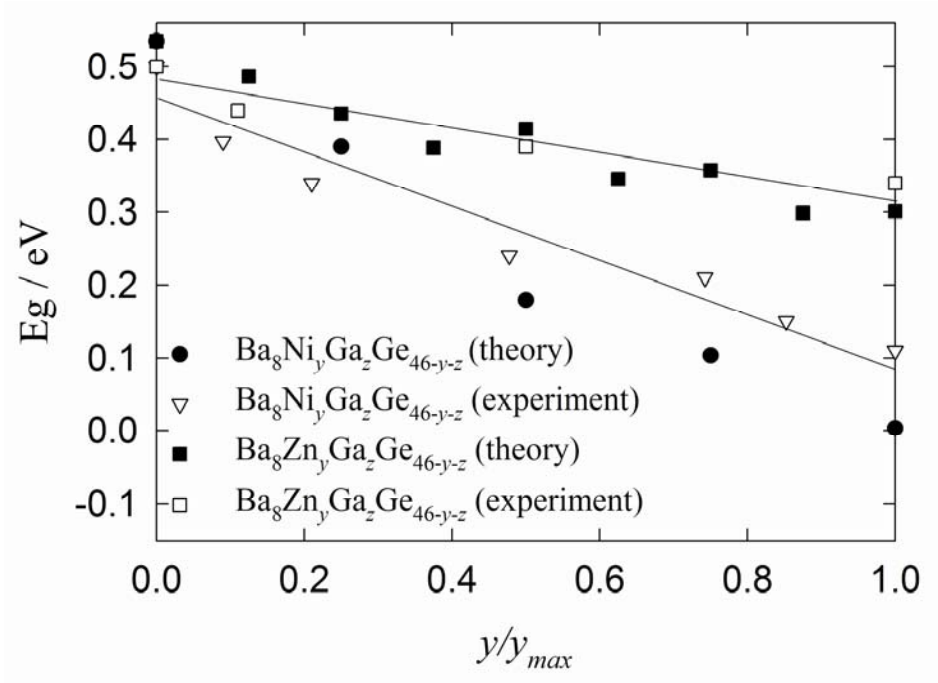
ULVAC ZEM-3 for electrical resistivity and Seebeck coefficient

Anter flash diffusivity system for thermal conductivity measurements

Utilize the leading-edge capabilities at the HTML User Program to determine reliable thermophysical and transport properties for thermoelectric materials from 300K to 800K.

General Motors R&D Center User Project: Accomplishments

- GM designed clathrates with composition that was changed by systematic cross-substitution of elements in the framework structure.
- Experimental measurements verified the ability to "tune" the band gap between 0.1eV and 0.5eV.

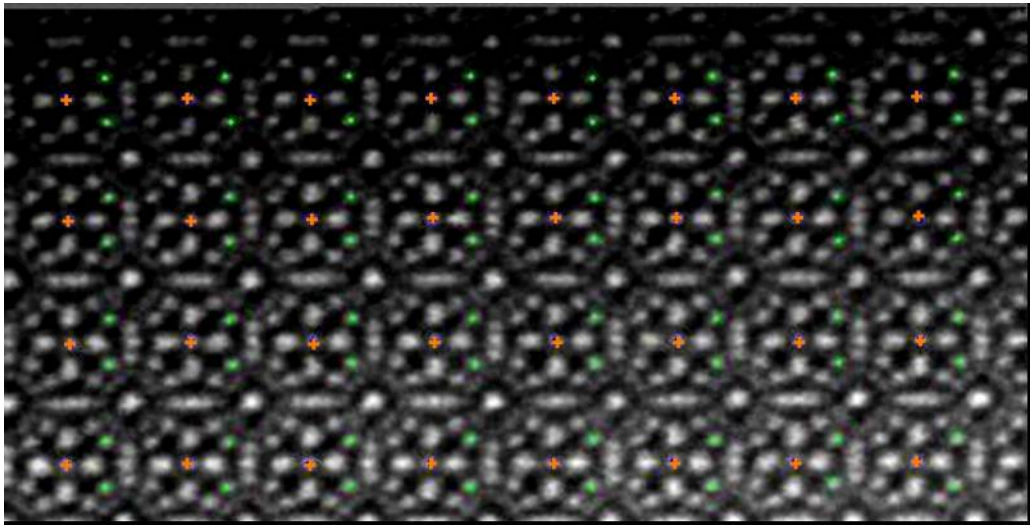


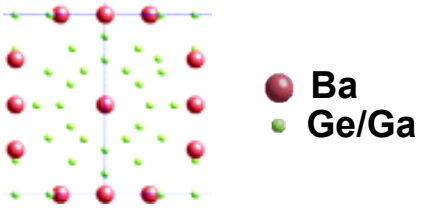
Dr. Xun Shi from General Motors analyzes transport measurement data.

General Motors R&D Center User Project: Accomplishments

Images taken using the HTML's aberration-corrected scanning/transmission electron microscope (STEM) along the [100] direction for $Ba_8Ni_yGa_zGe_{46-y-z}$ confirmed:

- *defect-free grains with no secondary phases*
- *intensity ratio per atom between 6c and 16i (or 24k) site is 0.93*
- *an element substitution is preferred at 6c sites*

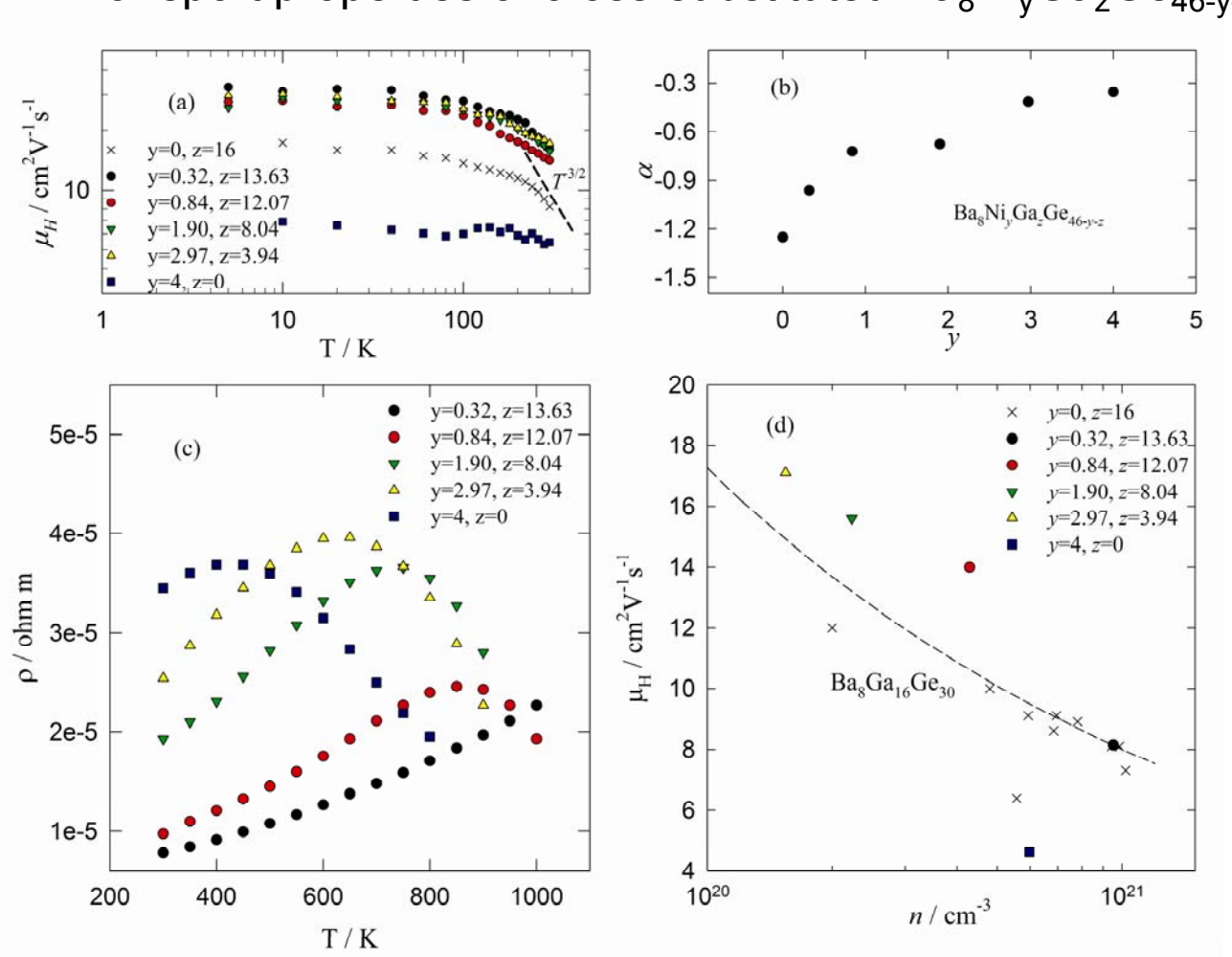




Orange dots represent 6c sites while green dots represent 16i or 24k sites.

General Motors R&D Center User Project: Accomplishments

Transport properties of cross-substituted $Ba_8Ni_yGa_zGe_{46-y-z}$ clathrates



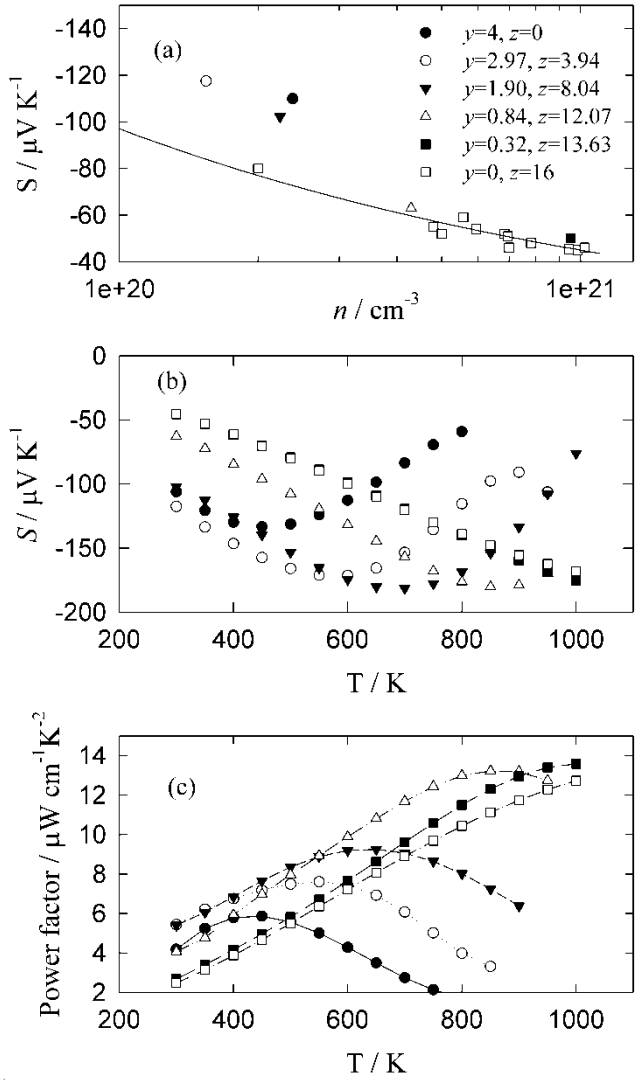
(a) temperature dependence of carrier mobility,

(b) room temperature carrier scattering parameter, *α*, as a function of y,

(c) temperature dependence of electrical resistivity,

(d) room temperature carrier mobility as a function of electron density.

General Motors R&D Center User Project: Accomplishments



High-efficiency $Ba_8Ni_yGa_zGe_{46-y-z}$ clathrate thermoelectric materials developed by General Motors and characterized at the HTML:

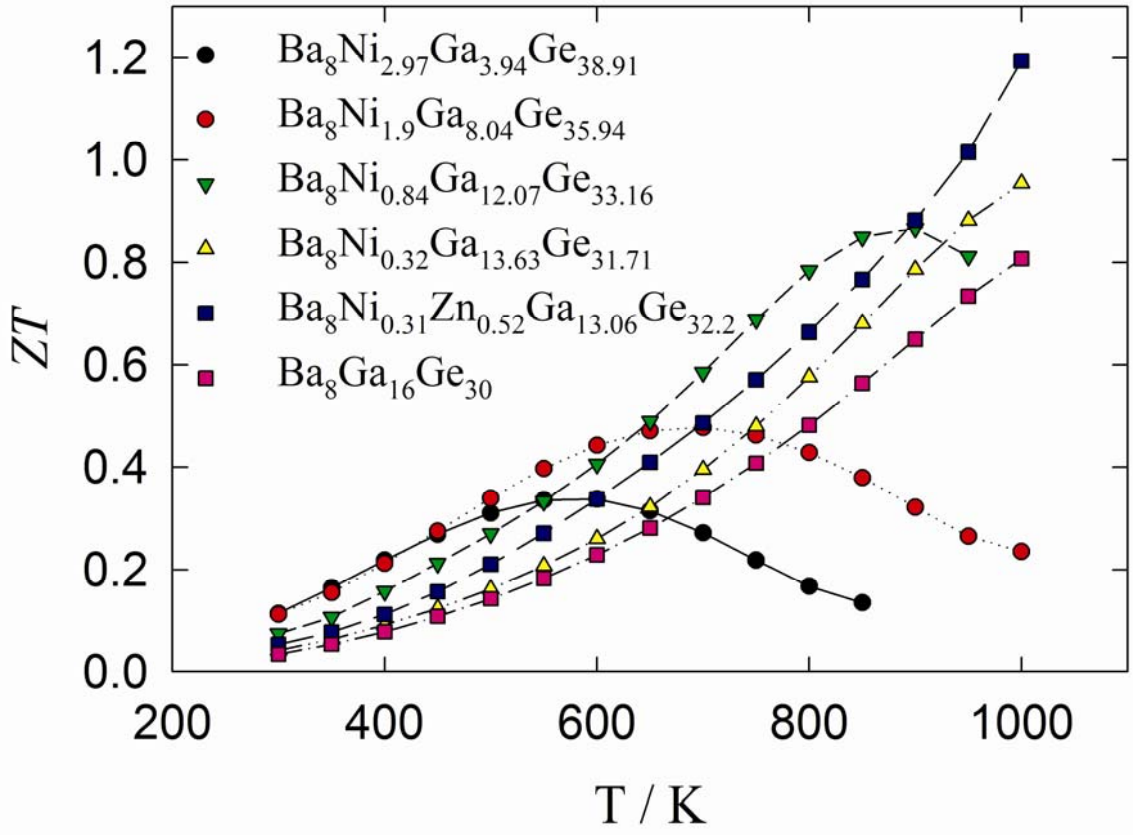
(a) room temperature thermopower as a function of electron density

(b) temperature dependence of thermopower *S*

(c) temperature dependence of power factor $S^2\sigma$.

General Motors R&D Center User Project: Accomplishments

Temperature dependence of *ZT* for transition metal-substituted $Ba_8Ga_{16}Ge_{30}$. Maximum ZT is 1.2 near 1000K.



General Motors R&D Center User Project: Future Work

GM plans to continue its collaboration with the HTML through new projects submitted to the HTML User Program, which will provide GM researchers with access to world-class expertise and characterization tools and achieve project goals set by the DOE Office of Vehicle Technologies.

General Motors R&D Center User Project: Summary

- GM utilized the world-class characterization capabilities available through the HTML User Program to determine the thermophysical and transport properties of cross-substituted clathrate thermoelectrics with a wide range of compositions developed by General Motors.
- The use of atomic-scale resolution STEM at the HTML enabled the establishment of fundamental property-structure relationships for clathrate thermoelectrics.
- The results obtained through this HTML User program project has contributed towards achieving the goals of the GM team to develop waste heat thermoelectric generators.