ANNOUNCEMENT

PART I: STI PRODUCT DESCRIPTION (To be completed by Recipient/Contractor)

A. STI Product Identifiers

1. REPORT/PRODUCT NUMBER(s)
Final Technical Report

2. DOE AWARD/CONTRACT NUMBER(s)
DE-FC26-04NT42278

3. OTHER IDENTIFYING NUMBER(s)

B. Recipient/Contractor

General Motors LLC

H. Sponsoring DOE Program Office

EERE Vehicle Technology

I. Subject Categories (list primary one first)

30 DIRECT ENERGY CONVERSION

Keywords

J. Description/Abstract

We conducted a vehicle analysis to assess the feasibility of thermoelectric technology for waste heat recovery and conversion to useful electrical power and found that eliminating the 500 W of electrical power generated by the alternator corresponded to about a 7% increase in fuel economy (FE) for a small car and about 6% for a full size truck. Electric power targets of 300 W were established for city and highway driving cycles for this project. We obtained critical vehicle level information for these driving cycles that enabled a high-level design and performance analysis of radiator and exhaust gas thermoelectric subsystems for several potential vehicle platforms, and we identified the location and geometric envelopes of the radiator and exhaust gas thermoelectric subsystems. Based on this analysis, we selected the Chevrolet Suburban as the most suitable demonstration vehicle for this project. Our modeling and thermal analysis assessment of a radiator-based thermoelectric generator (TEG), however, revealed severe practical limitations. Specifically the small temperature difference of 100°C or less between the engine coolant and ambient air results in a low Carnot conversion efficiency, and thermal resistance associated with air convection would reduce this conversion efficiency even further. We therefore decided not to pursue a radiator-based waste heat recovery system and focused only on the exhaust gas. Our overall approach was to combine science and engineering: (1) existing and newly developed TE materials were carefully selected and characterized by the material researcher members of our team, and most of the material property results were validated by our research partners, and (2) system engineers worked closely with vehicle engineers to ensure that accurate vehicle-level information was used for developing subsystem models and designs, and the subsystem output was analyzed for potential fuel economy gains. We incorporated material, module, subsystem, and integration costs into the material selection criteria in order to balance various materials, module and subsystem design, and vehicle integration options. Our work on advanced TE materials development and on TEG system design, assembly, vehicle integration, and testing proceeded in parallel efforts. Results from our two preliminary prototype TEGs using only Bi-Te TE modules allowed us to solve various mechanical challenges and to finalize and fine tune aspects of the design and implementation. Our materials research effort led us to quickly abandon work on PbTe and focus on the skutterudite materials due to their superior mechanical performance and suitability at automotive exhaust gas operating temperatures. We synthesized a

sufficiently large quantity of skutterudite material for module fabrication for our third and final prototype. Our TEG#3 is the first of its kind to contain state-of-the-art skutterudite-based TE modules to be installed and tested on a production vehicle. The design, which consisted of 24 skutterudite modules and 18 Bi-Te modules, attempted to optimize electrical power generation by using these two kinds of TE modules that have their peak performance temperatures matched to the actual temperature profile of the TEG during operation. The performance of TEG#3 was limited by the maximum temperature allowable for the Bi-Te TE modules located in the colder end of the TEG, resulting in the operating temperature for the skutterudite modules to be considerably below optimum. We measured the power output for (1) the complete TEG (25 Watts) and (2) an individual TE module series string (1/3 of the TEG) operated at a 60°C higher temperature (19 Watts). We estimate that under optimum operating temperature conditions, TEG#3 will generate about 235 Watts. With additional improvements in thermal and electrical interfaces, temperature homogeneity, and power conditioning, we estimate TEG#3 could deliver a power output of about 425 Watts.

C. STI Product Title

Program Final Report - Develop Thermoelectric Technology for Automotive Waste Heat Recovery

D. Author(s)

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E. STI Product Issue Date/Date of Publication

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L. Recipient/Contractor Point of Contact Contact

for additional information (contact or organization name to be included in published citations and who would receive any external questions about the content of the STI Product or the research contained therein)

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<u>05/23/2005</u>	<i>Thru</i>	<u>10/31/2011</u>		<u>General Motors LLC</u>
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**Program Final Report - Develop Thermoelectric
Technology for Automotive Waste Heat Recovery
Period Covered: May 23, 2005 through October 31, 2011
Date of report: January 29, 2012**

Prepared by:

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U.S. Department of Energy
National Energy Technology Laboratory

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EXECUTIVE SUMMARY

This cooperative development project develops thermoelectric (TE) technology for automotive waste heat recovery. The National Energy Technology Laboratory awarded General Motors (GM) a Cooperative Agreement on August 19, 2004. This Project Final Report details work performed under this program between May 23, 2005 and October 31, 2011.

We conducted a vehicle analysis to assess the feasibility of thermoelectric technology for waste heat recovery and conversion to useful electrical power and found that eliminating the 500 W of electrical power generated by the alternator corresponded to about a 7 % increase in fuel economy (FE) for a small car and about 6% for a full size truck. Electric power targets of 300 W were established for city and highway driving cycles for this project. We obtained critical vehicle level information for these driving cycles that enabled a high level design and performance analysis of radiator and exhaust gas thermoelectric subsystems for several potential vehicle platforms, and we identified the location and geometric envelopes of the radiator and exhaust gas thermoelectric subsystems. Based on this analysis, we selected the Chevrolet Suburban as the most suitable demonstration vehicle for this project. Our modeling and thermal analysis assessment of a radiator-based thermoelectric generator (TEG), however, revealed severe practical limitations. Specifically the small temperature difference of 100°C or less between the engine coolant and ambient air results in a low Carnot conversion efficiency, and thermal resistance associated with air convection would reduce this conversion efficiency even further. We therefore decided not to pursue a radiator-based waste heat recovery system and focused only on the exhaust gas. Our overall approach was to combine science and engineering: (1) existing and newly developed TE materials were carefully selected and characterized by the material researcher members of our team, and most of the material property results were validated by our research partners, and (2) system engineers worked closely with vehicle engineers to ensure that accurate vehicle-level information was used for developing subsystem models and designs, and the subsystem output was analyzed for potential fuel economy gains. We incorporated material, module, subsystem, and integration costs into the material selection criteria in order to balance various materials, module and subsystem design, and vehicle integration options. Our work on advanced TE materials development and on TEG system design, assembly, vehicle integration, and testing proceeded in parallel efforts. Results from our two preliminary prototype TEGs using only Bi-Te TE modules allowed us to solve various mechanical challenges and to finalize and fine tune aspects of the design and implementation. Our materials research effort led us to quickly abandon work on PbTe and focus on the skutterudite materials due to their superior mechanical performance and suitability at automotive exhaust gas operating temperatures. We synthesized a sufficiently large quantity of skutterudite material for module fabrication for our third and final prototype. Our TEG#3 is the first of its kind to contain state-of-the-art skutterudite-based TE modules to be installed and tested on a production vehicle. The design, which consisted of 24 skutterudite modules and 18 Bi-Te modules, attempted to optimize electrical power generation by using these two kinds of TE modules that have their peak performance temperatures matched to the actual temperature profile of the TEG during operation. The performance of TEG#3 was limited by the maximum temperature allowable for the Bi-Te TE modules located in the colder end of the TEG, resulting in the operating temperature for the skutterudite modules to be considerably below optimum. We measured the power output for (1) the complete TEG (25 Watts) and (2) an individual TE module series string (1/3 of the TEG) operated at a 60°C higher temperature (19 Watts). We estimate that under optimum operating temperature conditions, TEG#3 will generate about 235 Watts. With additional improvements in thermal and electrical interfaces, temperature homogeneity, and power conditioning, we estimate TEG#3 could deliver a power output of about 425 Watts.

1. ANALYSIS AND SELECTION OF THE DEMONSTRATION VEHICLE FOR A PROTOTYPE THERMOELECTRIC GENERATOR

The kickoff meeting for the GM-led project to develop advanced thermoelectric (TE) technology for automotive waste heat recovery was held on May 23, 2005, at the DOE headquarters in Washington, DC. The first task of the project was to conduct vehicle analysis to assess the feasibility of thermoelectric technology for waste heat recovery and conversion to useful electrical power. Vehicle-level fuel economy studies were carried out using GM's Overdrive simulation tool, which solves systems of ordinary differential equations that approximate the rigid body dynamics of the vehicle. Overdrive has been used for many of GM's past and future vehicle development cycles. The output from Overdrive was a set of parameters that reflect vehicle performance, fuel economy, drive quality, and energy management. Figure 1 shows the dependence of fuel economy (FE) on electrical load (EL) at the alternator calculated using Overdrive for a group of representative cars and trucks. From this, we concluded that eliminating the 500 W of electrical power generated by the alternator corresponded to about a 7 % increase in FE for a small car and about 6% for a full size truck. According to the EPA's Federal Test Procedure (FTP), however, a typical vehicle EL is about 300 W.

Figure 1. Dependence of fuel economy (FE) on electrical load (EL) at the alternator for representative cars and trucks

To analyze the full impact of a thermoelectric generator (TEG) on FE, we included the increase in the overall vehicle weight due to the TEG itself. Figure 2 shows the dependence of FE on EL at the alternator for a midsize truck at various vehicle weights. For a weight of 4417 lbs (2004 kg) and EL = 300 W, the EPA composite FE rating is 18.6 mpg. A 10% FE improvement means an FE increase of $\Delta \text{mpg} = 1.86$ mpg, and based on our analysis, the mass penalty target for the TEG should not exceed 5% of this 1.86 mpg gain. The data in Fig. 2 for the midsize truck indicate the rate of decrease in FE

improvement with increasing mass equals $(18.81 \text{ mpg} - 18.39 \text{ mpg}) / (4667 \text{ lbs} - 4167 \text{ lbs}) = 1 \Delta \text{mpg} / 1190 \text{ lbs} = 1 \Delta \text{mpg} / 540 \text{ kg} = 1.85 \Delta \text{mpg} / 1000 \text{ kg}$. Therefore, the total mass of a TEG system for a midsize truck should be less than $(5\%) \times (1.86 \Delta \text{mpg}) / (1.85 \Delta \text{mpg} / 1000 \text{ kg}) = 50 \text{ kg}$. We also observe from Fig. 2 that reducing the vehicle EL from 300 W to zero using a TEG would only improve FE by about 0.6 mpg, or about 3%, for the FTP drive cycle. Consequently, to reach the DOE goal of 10% FE improvement, more electrical power usage by the vehicle electrical system must be displaced by TE generated electrical power during the test drive cycle. This means additional vehicle electrification, which could be accomplished by shifting some of the engine-driven accessories to electrical drive in order to raise the electrical accessory load consumption. Examples of this include electric power steering, an electric coolant pump, an electric cooling fan, oil pump assist, electric valve actuation and timing, and electric heating for catalytic converters. The thermoelectrically generated electrical power could also be used directly for propulsion, which has the potential for the largest FE gain and is most easily accomplished in a hybrid vehicle.

Figure 2. Dependence of FE on EL at the alternator for a midsize truck at various weights for an FTP drive cycle. Symbols are data generated by Overdrive, and lines are guides for the eye.

In selecting a suitable vehicle for TE power generation from automotive waste heat, we considered both the radiator and exhaust gas as potential low and high temperature heat sources, respectively. Additional considerations for the selection of a demonstration vehicle for this project were (1) that the coolant and exhaust gas heat and mass flow were sufficient to support TE power generation, (2) that the vehicle's coolant system was adequate to cool the cold side of the exhaust gas TEG without any additional coolant pump or radiator capacity with the added heat load from the exhaust gas based TEG, (3) that the space available for the TEG was sufficiently large to accommodate a reasonable design for both radiator- and exhaust gas-based TEGs, and (4) that the necessary vehicle modifications to the

existing production vehicle were relatively simple and consistent with the scope of this project. We focused on assessing the economic feasibility of automotive TE waste heat recovery, selecting candidate TE materials for radiator and exhaust gas heat recovery modules, and analyzing the design and performance of radiator and exhaust gas heat recovery subsystems. As part of the economic feasibility study, we completed cost and manufacturability assessments for thin-film (for a radiator TEG) and bulk (for an exhaust gas TEG) thermoelectric devices. Electric power targets of an average of 300 W were established for city and highway driving cycles. We obtained critical vehicle level information for these driving cycles that enabled a high level design and performance analysis of radiator and exhaust gas thermoelectric subsystems for potential vehicle platforms. We also identified the location and geometric envelopes of the radiator and exhaust gas thermoelectric subsystems. We then selected the Chevrolet Suburban as the most suitable demonstration vehicle for this project.

We next focused on the viability of automotive TE waste heat recovery subsystem based on radiator and exhaust gas heat of the Chevrolet Suburban. This included specific design and performance estimates of the radiator and exhaust gas heat recovery subsystems using vehicle performance data for FTP city and highway driving cycles such as temperatures, heat flow rates, air flow rates, and exhaust gas chemistries. Our modeling and thermal analysis assessment of a radiator-based TE generator, however, revealed severe practical limitations. The small temperature difference of $\Delta T < 100^{\circ}\text{C}$ between the engine coolant (hot side) and ambient air (cold side) results in a low Carnot conversion efficiency, and thermal resistance associated with air convection would reduce this conversion efficiency even further. In addition, the heat release requirements of the vehicle coolant system via the radiator do not match the heat flow conditions of the TEG for optimum electrical power generation without significantly increasing the surface area of the radiator, which we considered impractical. Finally, the coolant flow through the radiator that determines the cold side temperature is only intermittent during the FTP drive cycle, and this reduces the time-averaged TE power generation. These results plus cost considerations eventually led us to abandon further work on a radiator-based TEG. We then focused on the exhaust gas heat source where the higher temperatures lead to a higher heat to electricity conversion efficiency and a more compact generator design.

1. THERMOELECTRIC GENERATOR SYSTEM DESIGN

For the design and analysis of the exhaust gas TEG system, we developed two primary steady-state thermal models to solve a large system of non-linear equations to assess the steady-state power output for a given exhaust flow condition. The first model was a bottoms-up analysis using the TE properties of each n-type and p-type leg material, the specific leg packing arrangement, the properties of the electrical conductors, and thermal and electrical contact resistances at each interface. This model was used to assess and optimize the generator design in lieu of module properties. The second model was

developed using the TE modules performance of commercial Bi-Te modules and idealized PbTe modules and allows for two different types of modules to be used simultaneously in the TEG. Our design concept was to use higher temperature PbTe (and eventually skutterudite) modules in the inlet (hotter) section of the TEG and lower temperature Bi-Te modules in the downstream (cooler) section. Four types of commercial Bi-Te modules and two PbTe modules with assumed properties were incorporated into the model, as well as varying numbers of modules in each section. We also developed several analysis tools for simulating TEG transient performance and average performance over a drive cycle, and these tools were verified with more accurate and computationally intense transient 3D thermoelectric finite element models. We further developed the transient modeling capability building a MATLAB Simulink model of the vehicle exhaust system, TE generator, and bypass controller as lumped-elements that represent the system as thermal masses and capacitances. The Simulink model determined the transient power output by analyzing the exhaust gas flow conditions and controlling the exhaust gas bypass valve to protect the TEG from excessive temperature or back pressure. Refinement of the electrical, thermal, and mechanical aspects of the TEG design was accomplished via the simulation and modeling work. Connection schemes for the TE modules were evaluated for optimum electrical power generation, and the number, type, and arrangement of TE modules were defined along with the mechanical design of the hot- and cold-side heat exchangers based on the physical constraints determined by the demonstration vehicle exhaust system and an overall cost metric $\$/W$. This metric included estimates of TE material, fabrication, and assembly costs. The steps in the TEG design process is shown schematically in Fig. 3

Figure 3. Steps in the design process for the TEG. This included optimizing (a) the TE module size and shape; (b) the heat exchanger aspect ratio, TE module placement on the heat exchanger, and mechanical components; and (c) the final TEG design based on a two-stage TE module arrangement, thermal and mechanical constraints, and vehicle integration requirements. Refinement of the final design included the program metric ($\$/Watt$) to maximize electrical power for the lowest estimated cost.

Special attention was placed on ensuring that the TEG parts were machinable and could actually be assembled. Mechanical compatibility with the various automotive components was also incorporated into the final design as was flexibility for swapping out TE modules in case of failure or TE module upgrades. Significant effort was expended on the thermo-mechanical design, machining, fabrication, and assembly of the TEG, which included mitigating the significant differences in thermal expansion between the heat exchangers and the TE module interfaces. Electrical components and instrumentation for integrating the TEG into the vehicle system and for TEG control and performance testing were installed in the vehicle, and the vehicle exhaust system was modified to include the bypass flow path, valves, and a controller that was needed to ensure the TEG remained within temperature and backpressure operating limits. Figure 4 shows the location of the TEG and some of the subsystems on the demo vehicle.

Figure 4. Location of the prototype TEG on the Chevrolet Suburban demonstration vehicle.

The overall approach we used to achieve the project goal was to combine science and engineering. Existing and newly developed materials were carefully selected and characterized by the material researcher members of our team, and most of the material property results were validated by our research partners. System engineers worked closely with vehicle engineers to ensure that accurate vehicle level information was used for developing subsystem models and designs, and the subsystem output was analyzed for potential fuel economy gains. We incorporated material, module, subsystem, and integration costs into the material selection criteria in order to balance various materials, module and subsystem design, and vehicle integration options. Based on our TE module, heat exchanger, and subsystem design,

our modeling and simulation results showed that the average electrical power output over the FTP city cycle of an exhaust waste recovery system would need to be higher than the 350 W minimum for a calculated 3% fuel economy improvement, and this indicated that a more aggressive test cycle, such as the US06, would be more appropriate to assess TEG performance.

We developed a dc-to-dc power conversion strategy that included component definition and electrical circuit simulation modeling to examine and optimize high-level TEG output performance. We assessed the lumped element electric properties and impedance of a typical commercial TE module, and the results indicated that the low capacitance and inductance of the module should have no effect on the optimum power-tracking scheme. A simplified transient model was implemented in Matlab Simulink to simulate the dynamic performance of the TE generator system. A proportional controller was designed and simulated in the Simulink model to control the exhaust bypass valve to ensure that the TEG remained within temperature and backpressure service limits.

2. THERMOELECTRIC MATERIAL RESEARCH

Throughout this project, we extensively explored many new TE materials and conducted a detailed optimization of several existing state-of-the-art TE materials. On the theoretical side, we conducted a systematic computational study of the thermodynamic properties of new and promising thermoelectric materials. We made significant progress in the development of a computational approach to calculating lattice thermal conductivity based on the Boltzmann transport equation and first-principles density functional theory (DFT) calculation, although more work is needed. We calculated the phonon density of states (DOS) for LAST-m and AgSbTe_2 over a range of doping concentrations using a very large 1000-atom unit cell, and our results indicate that the response of electronic DOS near the Fermi energy in LAST-m is markedly different from those obtained by previously reported work where calculations have been limited to smaller super-cells. We performed a computational study of nano-cluster doping in several LAST-type materials to compare with the results of traditional impurity doping models and studied the energetics and nano-cluster-host interactions in LAST-m in order to reveal systematic structural evolution of AgSb nano-clusters in the PbTe host lattice. Additional calculations were carried out for AgSbTe_2 to examine its phonon density of states and thermodynamic properties for comparison to neutron scattering and transmission electron microscopy (TEM) experiments. Finally, we calculated the electronic band structure of NiSn Co-doped skutterudites to investigate the power factor enhancement seen in experiments.

On the experimental side, we measured the transport and thermoelectric properties of many new candidate TE materials. We identified a method of enhancing the TE figure of merit, ZT, in various doped clathrate compounds, and we determined the correlation between lattice thermal conductivity and structural properties of antiferrofluorites, potentially cost-effective TE materials. This work included an

investigation of diamond-like Cu-Ge-Se phases that have an unexpectedly low thermal conductivity at high temperatures. We measured the mechanical properties of TAGS materials and established a controllable p-type doping procedure for PbTe-based materials. The materials research team members continued the exploration of new compositions and processing conditions for optimization of existing TE materials, primarily skutterudites. Multiple filled skutterudites and non-equilibrium grown skutterudites were extensively studied and characterized. We were able to achieve $ZT = 1.6$ in the temperature range relevant to automotive exhaust TE waste heat recovery for multiple-filled n-type skutterudites, which was the highest ZT value observed for skutterudites at that time. High temperature mechanical property characterizations were also carried out for the various materials we considered for TE module fabrication. One of the main results from the material research part of this project was a demonstration that cost-effective skutterudite materials were very promising TE material candidates for our TEG prototype from the point of view of TE material performance, mechanical properties, module fabrication, and cost.

3. MATERIAL SELECTION FOR THERMOELECTRIC MODULE FABRICATION

For TE module development and production, we initially focused on the well-known PbTe-based materials. We synthesized several compositionally optimized ingots and processed a portion of those ingots into metal-coated elements for testing and evaluation prior to assembly into TE modules. We determined that thick metal plating could be made to adhere well to the PbTe materials, and the problems of braze wetting thermally sprayed metal diffusion barriers were solved. A direct bond aluminum ceramic option for providing electrical interconnects between TE elements was also developed. We then produced n-type and p-type PbTe-based ingots, fabricated TE modules, and subjected them to thermal cycle and aging tests. TE module internal electrical resistance was monitored after each thermal cycle and cross sections of the module were evaluated. Significant cracking and fractures occurred in both the p-type and n-type elements, and the propagation of these fractures caused an increase in device internal resistance during the thermal cycling test and ultimately resulted in severe degradation of TE module performance and open circuit failures. These results indicated that further development of PbTe-based TE modules would likely require considerable work, but we had no real confidence that suitably robust modules could be obtained during this project. With considerable progress on optimizing and developing skutterudites as an alternative to PbTe, we decided to switch to the more mechanically robust skutterudite materials for continued TE module development.

Initial work on skutterudites consisted of testing a module assembly technique for bonding the p- and n-type skutterudite elements to an electrical interconnect tab using a combination of braze and flux. Results were very promising, particularly in comparison to similarly processed PbTe thermoelectric elements where the significant cracking occurred after processing and thermal cycling. Specimens of p-type and n-type skutterudites were subjected to a heat treatment in air to evaluate their physical/chemical

stability under these conditions. Severe degradation was observed, particularly for p-type material, shown in Fig. 5, and these results dramatically demonstrated the need for the skutterudite modules to be protected by an inert environment at operating temperatures to prevent rapid degradation and failure. Our TEG system design anticipated this need, by including a hermetically sealed aluminum box surrounding the assembled TE modules, heat exchangers, and other components. In addition, we investigated the feasibility of encapsulating the individual TE legs and modules as a more cost effective solution to the problem of oxidation and sublimation of skutterudite materials at elevated temperatures.

Figure 5. Images of p-type skutterudite after heating to 450°C in air.

A process for fabricating high temperature skutterudite-based TE modules using a controlled atmosphere furnace brazing (CAB) technique was developed that had the advantages of high volume throughput and low cost. Both p-type skutterudite legs (with old diffusion barrier processing parameters) and n-type legs (with new diffusion barrier processing parameters), shown in Fig. 6, were successfully fabricated. Next, a large volume of skutterudite material was synthesized for TE module fabrication using conventional melting, grinding, sintering, and annealing techniques in order to get the correct phase with optimum compositions and transport properties. Final billets were formed using spark plasma sintering (SPS) processing. Considerable work was needed for the SPS step to ensure proper consolidation that avoided metallurgical flaws and compositional inhomogeneities that otherwise would degrade performance of both the thermoelectric figure of merit ZT and the mechanical strength. More details of the skutterudite material synthesis and module fabrication will be discussed below in Section 6. While this skutterudite material development work for TE modules was in progress, we proceeded to fabricate, assemble, and test two prototype TEGs.

4. TE GENERATOR PROTOTYPE PRELIMINARY TESTING: TEG#1 & TEG#2

The first assembly of our prototype generator, TEG#1, was completed using only three Bi-Te-based TE modules. It was installed and operated on the Chevrolet Suburban test vehicle for preliminary validation and testing of the various control and integration subsystems. We found that the first control unit for the bypass valve failed and had to be replaced, after which both the bypass valve control and the vehicle data collection systems were fully functional and operating correctly. We measured the

temperature profile and TE module output performance. The TEG temperature as measured by the thermocouple on the hot-side heat sink at the exhaust gas input (i.e., front and center position as shown in Fig. 7) could then be controlled to a programmed set point. Figure 8 shows the temperature profile of this front and center thermocouple attached to the heat exchanger along with the temperatures of the input exhaust gas as it enters the TEG, the output exhaust gas, and the engine coolant used in the TEG cooling blocks. The programmed set point for the maximum heat exchanger temperature was 250°C, but as expected, a substantial temperature drop was observed along the length of the TEG, falling to 178°C and 148°C in the middle and rear of the heat exchanger, respectively, as shown in Fig. 9. On the other hand, we found that the temperature variation across the TEG was small, less than about 3°C, also shown in Fig. 9. The output open circuit voltages of the three TE modules located at the front, center, and rear of the TEG exhibited values consistent with a temperature difference (ΔT) across the TE modules of about 50°C less than the measured temperature difference between the heat exchanger and coolant.

Figure 6. Skutterudite TE legs with diffusion barriers. The three on the left are p-type, and the three on the right are n-type.

TEG#1 was installed on the demonstration vehicle for about 30 days and subjected to approximately 50 cold starts during that time; the total time of vehicle operation was approximately 7 hours. The maximum temperature excursion of the heat exchanger at the front TE module location was about 275°C for about half the time during operation, although the module itself is estimated to only reach around 240°C. The heat exchanger reached 300°C on two occasions. A challenge in assembling TEG#1 was making a structurally robust and gas-tight joint between the nickel-plated copper heat exchanger and the stainless steel exhaust gas spreaders. This was unsuccessful and resulted in exhaust gas leakage into the body of TEG#1. We found no measurable degradation of Bi-Te module performance during the vehicle test drives. Moreover, we examined the three Bi-Te modules after operation and found no visible degradation. The Bi-Te module performance at room temperature was measured using a Z-Meter (from RMT Ltd.) and these results also indicated no measurable degradation. In fact, the only evidence that TEG#1 had been in operation was a significant amount of condensed water inside the unit and the rust that developed on various exterior and interior iron (i.e., non-stainless steel) parts. Figure 10 shows various parts of the disassembled generator after many hours of vehicle driving.

Figure 7. Placement of thermocouples and the connection to one of the Bi-Te module in the Front and Center position of TEG#1. The square blocks connected by the small bellows make up the cold-side heat exchanger that is cooled by the engine coolant.

Figure 8. The input and output exhaust gas temperatures and the engine coolant temperature for TEG#1. Also shown are the Front and Center heat exchanger temperatures.

Figure 9. Temperature measurements at the Front, Middle, and Rear positions of the heat exchanger show a substantial temperature drop of nearly 100°C along the length of the heat exchanger. For each position, however, the temperature variation in the transverse direction, Center, Right, and Left, is only a few °C.

Figure 10. (a) Rusty clamp bolts resulting from exhaust gas infiltration into TEG#1 during vehicle drive testing, (b) the external end plate of the TEG case shows rust after exposure to the outside environment, and (c) the hot side nickel-plated copper heat exchanger shows negligible corrosion after operation. The white areas are thermal grease residue after removal of the Bi-Te modules and ceramic blank plates.

Next, a complete generator, TEG#2, was assembled with 42 Bi-Te modules installed on the heat exchanger, so adequate testing of the various components and subsystems, including the dc-to-dc converter and the vehicle integration and controls, could be completed prior to installing skutterudite TE modules into the final generator, i.e., TEG#3. A remaining difficulty with the assembly of TEG#2 was ensuring a complete and gas-tight seal in the joint between the stainless steel gas spreaders and the copper heat extractor. Small pinhole leaks remained that allowed a small amount of exhaust gas to infiltrate into the body of TEG#2 and prevented us from keeping the interior of the TEG filled with inert gas during operation. It was evident from the results of TEG#1 testing, where there was essentially no seal between the spreaders and the heat extractor, that the effect on the performance of the Bi-Te TE modules was not affected, and indeed, the remaining small leakage in TEG#2 did not degrade the performance of the Bi-Te modules. For TEG#3 with skutterudite TE modules, however, it was necessary to have gas tight joints and an inert gas environment to prevent rapid oxidation for the skutterudite TE material during operation.

In TEG#2, each row of seven TE modules along the length of the heat exchanger was connected into a series string as shown in Fig. 11. This way, each row would experience the same temperature profile from the inlet to outlet ends of the TEG, and hence the output voltage of each string should be approximately the same. This connection scheme also gave us flexibility in connecting the seven-module strings in various combinations of parallel and series arrangements for maximum performance. A set of circuit boards was fabricated for connecting each series string of modules, as shown in Fig. 12, and a diode was connected across each module at the circuit board to allow current to flow through the circuit in the event of an open circuit failure of any TE module. The six rows of seven-module strings were then connected into two series strings of three rows each. The two resulting series strings of twenty-one modules each were connected to feed-thru wires to bring the electrical power outside the TEG. These wires were connected to the dc-to-dc converter for integration into the electrical system of the vehicle. Installation of the external case and installation of the TEG onto the vehicle proved to be difficult due to the tendency of the wires to get abraded and/or become pinched inside the TEG. Ultimately, several rewirings were needed to eliminate the short circuits. Consequently, we designed and fabricated a new case to provide more room inside the TEG. This made the case assembly easier and less likely to cause internal short circuits between the module wires and the case.

TEG#2 provided output power for fully testing and optimizing the power conditioning electronics for incorporation of the TEG output power into the main vehicle electrical system. The first data collected using TEG#2, Fig. 13, shows the generation of over 25 W and confirmed our ability to maintain the heat exchanger temperature at the input end of the TEG at the requisite 250°C throughout the test drive cycle. As expected, there was considerably more heat available for recovery than we could utilize because of the temperature limitation set by using Bi-Te TE modules in the TEG and because of the

substantial temperature drop of the exhaust gas as it flows down the length of the TEG.

Figure 11. The wiring of TEG#2 is shown in progress. Each set of seven modules in rows along the length of the heat exchanger connected in series using circuit boards located on the sides of the unit. One string of series-connected modules is encircled by the dotted red line.

Figure 12. The circuit board assembly with seven modules connected (red and blue wires at the top). The output of each circuit board is connected to the others and then to the feed-thru connections by the heavy gauge white electrical output wires.

Data was collected for TEG#2 under various operating conditions. Figure 14 shows the results for a cold start test conducted for about 25 minutes. After about 6 minutes, the system had warmed up and reached a steady-state temperature profile. The voltage, current, and power were measured as the external load resistance provided by the dc-to-dc converter was varied. Figure 15 shows the data between about 20 and 25 minutes (shaded region in Fig. 14) that we used for further analysis. These data were

examined in more detail to determine the optimum current and voltage for maximum power delivery from the 42 Bi-Te TE modules. The temperature of the heat exchanger ranged from about 255°C at the hot end (Front) to about 165°C at the cool end (Rear), and the coolant temperature was about 90°C. Based on our results for TEG#1, we estimate that the corresponding maximum TE module temperatures and ΔT s for TEG#2 to be $T_{\text{front}} = 220^\circ\text{C}$, $T_{\text{rear}} = 135^\circ\text{C}$, $\Delta T_{\text{front}} = 130^\circ\text{C}$, $\Delta T_{\text{rear}} = 45^\circ\text{C}$. Averaging over this temperature profile, the maximum power generation would be 1 W/module, or about 40 W total for TEG#2. The output current and power of TEG#2 was analyzed as a function of the TEG voltage. We estimated that the optimum voltage for TEG#2 power output should be about 21.5 Volts, corresponding to 0.543 Amps and 11.7 W. We conclude that further optimization of the TEG system is needed, including optimizing the thermal profile of the heat exchanger and maximizing ΔT across the TE modules.

Figure 13. Results for TEG#2 with 42 Bi-Te modules connected in two parallel strings of 21 modules each. The output power (red curve) was measured with the TEG output connected to a dc-to-dc converter with an optimized load for maximum power conversion. Heat exchanger temperatures are the blue (Front) and black (Rear) curves, and the green curve is the coolant temperature.

A Federal Test Procedure (FTP) driving cycle was conducted, and the results are shown in Fig. 16. Data for temperatures of the exhaust gas in and out of the TEG, the vehicle speed, the bypass valve position, and the TEG power output were collected for a cycle drive time of about 23 minutes. The

vehicle speed, shown as the light blue line in Fig. 16, varied from zero (vehicle stopped, engine idle) to 90 kph, and averaged about 32 kph over the duration of the test. The bypass control algorithm resulted in the bypass valve position (dashed red line) being about 75% closed once the engine warmed up after about 3 minutes and tracked with the vehicle speed. The resulting temperature control of the hot side heat exchanger (purple line) at the set point temperature of 250°C was reasonably good. As seen previously for TEG#1, the temperature gradient along the length of the heat exchanger, shown by the purple, green, and dark blue lines, was about 100°C from inlet to outlet, and the coolant temperature (dotted black line) was about 95°C throughout the drive cycle. The TEG output power (brown line) approached 30 W.

Figure 14. Temperatures and TEG outputs for a cold start drive test of TEG#2. After about 6 minutes, the temperature profile approaches steady state values. Note that the data for Current has been multiplied by a factor of 10, i.e., the maximum current is about 0.9 Amp. The data between about 20 and 25 minutes (shaded region) is shown in Fig. 15.

Figure 15. Temperatures and TEG output as a function of time for a cold start drive test of TEG#2 on the Chevrolet Suburban demonstration vehicle between about 20 and 25 minutes after a cold start. Note that the data for Current has been multiplied by a factor of 10, i.e., the maximum current is about 0.9 Amp.

Figure 16. Results of an FTP drive cycle. Temperatures are shown for exhaust gas in (solid black line) and out (dashed black line) of the TEG, coolant (dotted black line), and Front (purple line), Middle (green line), and Rear (dark blue line) of the heat exchanger. The bypass valve position (dashed red line) varies inversely with vehicle speed (light blue), as expected, and results in relatively smooth heat exchanger temperatures. The TEG output power (brown line) approaches 30 W. The downward vertical spikes of (indicated as “Digital Noise” to the right of the figure) are associated with the power output optimizing algorithm for the dc-to-dc converter. The output power varies with heat exchanger temperature, which also tracks with vehicle speed, as expected. An enlargement of the data collected from 15 to 20 minutes is shown in Fig. 17.

An enlargement of the data collected from 15 to 20 minutes is shown in Fig. 17 that shows more explicitly how the widely varying vehicle speed (light blue line) results in large-scale temperature variation in the exhaust gas temperature (black line), and that the operation of the bypass valve (dashed red line) effectively smoothed out those temperature variations to give a more uniform temperature as a function of time (purple, green, and dark blue lines). It is clear that the better we can control the heat exchanger temperature profile during variations in vehicle engine output throughout the drive cycle, the more we can maximize TEG output power.

Figure 17. A portion of the FTP drive cycle showing detail for the various data signals for TEG#2. The digital noise in the output voltage (gold line) and power (brown line), which consisted of single data points periodically departing substantially from the curve (i.e., approximately 12 W and 12 V, respectively, lower than the curves shown) was removed.

5. SKUTTERUDITE MATERIALS FOR THERMOELECTRIC MODULE DEVELOPMENT

During the fabrication, assembly, and testing of TEG#1 and TEG#2, a parallel effort focused on developing, optimizing, and producing skutterudite materials for TE modules to be incorporated into the final generator, TEG#3. One goal was to develop a method for applying diffusion barriers on both n-type and p-type skutterudite materials, where good adhesion to the skutterudite material and metallurgical compatibility with the assembly brazes were key considerations during this work. As we scaled up the production of n-type skutterudites, we needed to ensure the transport properties remained acceptable. Thorough testing of thermal diffusivity, electrical resistivity, and Seebeck coefficient of typical specimens of n-type and p-type production skutterudite was performed and analyzed. The skutterudite materials were produced in the form of 30-mm diameter billets, as shown in Fig. 18, of which approximately half

were consumed for diffusion barrier development and improvements to materials synthesis processing conditions. In addition to excellent TE performance, both high fracture strength and resistance to thermal shock were needed to meet durability requirements for automotive waste heat recovery applications. Finite element analysis and fracture strength testing were performed, and the results, shown in Fig. 19, indicate that thermal tensile stresses are on the order of 130 MPa at a ΔT of 500°C; therefore, skutterudites can withstand stresses of over 140 MPa.

Figure 18. Billets of Skutterudite material ready to be sliced into parts for TE module fabrication.

Figure 19. Fracture strength tests indicate Skutterudites can withstand tensile stresses of over 140 MPa.

Because a major task in this project was the development of a metal contact layer for both the n- and p-type skutterudite materials, we focused on sputtering, electrolytic/electroless plating, and thermal arc spraying using selected metals. Adhesion was tested after application of the diffusion barrier and after 24-hour thermal treatment at 550°C. We identified two techniques that showed promise for successful adhesion. We developed a process for fabricating high-temperature TE modules using a controlled atmosphere furnace brazing technique. One of the main concerns was to minimize the temperature excursion of the TE material during the brazing cycle and to minimize the duration of the cycle. This reduced thermal stresses in the TEG module and minimized sublimation of volatile elements in the TE material, both of which would reduce thermoelectric performance. We fabricated several mini-TE modules for evaluating the integration of materials, diffusion barriers, braze joints, ceramics, etc., before scaling up the fabrication process.

The skutterudite materials with diffusion barriers were diced into TE legs and then attached to metalized ceramics using the braze method in a controlled atmosphere. After brazing, the top ceramic header was segmented using a high-speed diamond saw to keep the thermal stresses to a minimum when the TE module is subjected to large temperature gradients during operation. After fabrication, the mini-TE modules were thermally cycled and age tested to evaluate TE module integrity using measurement of the module internal ac resistance (ACR), which is the most sensitive indicator of TE device health. The results shown in Fig. 20 indicate that the device ACR for both types of skutterudite modules remained constant throughout the test, and this was a significant improvement compared to the PbTe device that exhibited ~18% increase in ACR under similar testing conditions, also shown in Fig. 20. The mini-TE modules were also cross-sectioned for microstructure examination. The TE modules made with direct-bonded alumina (DBA) ceramic performed the best and did not show any evidence of delamination. It was found that the skutterudite modules withstood the thermal cycling quite well, whereas the PbTe modules exhibited significant cracking in the PbTe material consistent with the increase in module impedance during the thermal cycling test. Additional mini-TE modules were fabricated and subjected to thermal age testing, and the results show that skutterudite modules were consistently more durable than the PbTe modules.

Based on these results for the mini-TE modules, we fabricated 29 full-scale, 50 mm x 50 mm, 32-uncouple skutterudite TE modules, shown in Fig. 21, for incorporation into the final TEG, TEG#3, for vehicle testing. We characterized these TE modules by measuring their impedance, and the results indicate a high degree of homogeneity and reproducibility in the material and fabrication process. The results also showed that porosity and reprocessing of the skutterudite billet material adversely affect the electrical resistance of the modules.

Figure 20. Comparison of temperature cycle test results for mini-TE modules made with skutterudites (green triangles and blue squares) and PbTe (red squares). The modules were cycled in N_2 , unclamped, in a belt furnace from room temperature up to 450°C for the skutterudites and up to 400°C for the PbTe modules. The duration of each cycle was ~20 minutes.

Figure 21. Full-scale skutterudite TE modules for assembly into TEG#3. These modules are 50mm x 50mm, and each has an array of 64 skutterudite legs (32 unicouples) bonded to ceramic plates.

Finally, the skutterudite modules were encapsulated using a low-cost, castable insulation material to prevent sublimation of antimony and increase efficiency by reducing parasitic heat loss. It is possible

that the encapsulation helped reduce oxidation, but we did provide an inert atmosphere established using a slight over-pressure of pure nitrogen gas inside the sealed case surrounding the TEG internal components.

6. TEG#3 ASSEMBLY AND TESTING

For the assembly of our final TEG, 24 skutterudite TE modules were incorporated into the hot (Front) end of the unit. The remaining 18 modules, located at the cooler end of the heat exchanger, were Bi-Te because of their better TE performance at the expected lower temperature in that portion of the heat exchanger. The heat exchanger assembly for TEG#3 is shown in Fig. 22. By making the heat exchanger out of stainless steel, we were able to make a significantly better gas-tight and structurally robust joint between the gas spreaders and the heat exchanger. In addition to the joining, the heat exchanger itself could be made with half the wall thickness, resulting in a lighter and much less expensive part.

Figure 22. Pictures of the welded stainless steel hot-side heat exchanger and gas spreader for TEG#3. This assembly prevents exhaust gas leakage into the body of TEG#3. This is a significant improvement over TEG#2, where the heat exchanger was fabricated out of copper and was therefore extremely difficult to join to the stainless steel gas spreader.

Figure 23 shows the layout of the upper side of the heat exchanger, including the thermocouple placement along the center row of TE modules. As in our previous TEG prototypes, the cooling blocks were connected by small bellows and joined together at the ends of the unit by coolant manifolds. In addition to the seven thermocouples shown in Fig. 23, another was installed at the front and middle left module positions. This allowed us to measure the lateral temperature variation during operation. In contrast to the previous TEG prototypes, which used a copper hot-side heat exchanger and stainless steel exhaust gas spreaders, TEG#3 used stainless steel for both the heat exchanger and spreaders.

Figure 23. The layout of TE modules for TEG#3 consisted of 24 skutterudite modules (Rows 1 – 4) and 18 Bi-Te modules (Rows 5 – 7) in two sections as described in the text. The positions labeled 1 – 4 correspond to the schematic drawings in Fig. 4 of the layers of copper and grafoil that make up the thermal interface between the heat exchanger and the skutterudite TE module. A similar set of interface layers were used for the Bi-Te modules in Rows 5 – 7. For each row, the same number of copper and grafoil layers was used for each module.

The placement of thermocouples, shown in Fig. 23, was intended for monitoring the temperature profile along the length of the heat exchanger. In order to maximize the output of the TEG, each thermocouple was monitored and used in the control algorithm for the bypass valve. For TEG#2, the thermocouples were inserted into small holes drilled into the surface of the hot side heat exchanger, and this gave very accurate readings of the heat exchanger temperatures in the front, middle, and rear positions, as well as the left, right, and center. For TEG#3, on the other hand, we decided on a different approach designed to get a better measure of the actual temperature of the hot side of the TE modules

along the length of the TEG. Each thermocouple was inserted into the crease of a folded rectangular piece of copper foil, as shown in the upper panel of Fig. 24. The dimensions of the foil were chosen so that it matched those of the hot side of the TE module when folded. This resulted in a minimum of two layers of copper foil between the TE module and the hot side heat exchanger.

Figure 24. Thermocouple installation for TEG#3. Thermocouples were inserted into a fold in a sheet of copper foil to ensure good thermal contact (upper panel). The foil was sandwiched between the hot-side of the TE modules and the heat exchanger surface. The lower panel shows more detail of the thermocouple-copper foil assembly inserted between the TE modules (which are under the aluminum cooling blocks) and the heat exchanger surface.

Additional layers of copper foil and grafoil were inserted between the folded copper foil and heat exchanger to build up additional thermal interfaces between the TE module and heat exchanger. This resulted in a reduction of the strongly varying temperature gradient along the length of the heat exchanger to give a more uniform temperature profile at the hot sides of the TE modules. All of the thermocouples were monitored, and the maximum temperature measured during the drive cycle was used for controlling

the bypass valve. This thermal profile scheme was implemented separately for the skutterudite and Bi-Te sections of the TEG, and there were separate upper-limit set point temperatures for each section. The lower panel of Fig. 24 shows in more detail the installation of the pieces of folded copper foil with inserted thermocouples. Our expectation was that we would be able to maintain a larger ΔT across the TE modules, but there still would be some temperature drop between the thermocouples and the TE module due to the actual properties of the thermal interfaces. Similarly, there would be a higher temperature on the cold side of the TE modules than measured by the coolant temperature due to the properties of the thermal interfaces between the cold side of the TE modules and the aluminum cooling blocks. Here, on the cold-side heat exchanger, we used the same thermal grease as we did for TEG#2, so we expect only a slightly larger temperature difference across the cold side interface than the 15°C observed for TEG#2. Finally, as in TEG#2, the TE modules were all connected via circuit boards with diodes in parallel for protection against the development of an open circuit. Each row of seven modules (4 skutterudite and 3 Bi-Te) were connected in series on one circuit board, and the output electrical power was conducted outside the TEG to the vehicle electrical system via the heavy-gauge wires, as shown in Fig. 25.

Figure 25. Assembly of TEG#3. All the cooling blocks are held in place using steel clamps transverse to the unit. The yellow circuit boards are shown at the side of the heat exchanger, and the thermocouple connections are arranged on top of the cooling blocks and between the clamps. The force exerted by the clamps on the cooling blocks and in turn on the TE modules and interfaces is adjusted by the nut and bolt at each end of the clamp.

As described above, the layout of TE modules in TEG#3 consisted of 24 skutterudite modules positioned at the hotter end of the heat exchanger adjacent to the exhaust gas inlet, and 18 Bi-Te modules positioned at the colder end of the heat exchanger adjacent to exhaust gas outlet. This arrangement was adopted in an attempt to take advantage of the substantial temperature drop of about 100°C along the length of the TEG resulting from the large heat flow from the exhaust gas into the heat exchanger, through the TE modules, into the cold side heat exchanger, and into the engine coolant. This heat flow, which was the energy stream used to generate electrical power by the TE modules, caused the exhaust gas temperature to decrease as it flowed through the TEG and resulted in a decreasing temperature profile along the length of the heat exchanger. To compensate for this temperature profile experienced by the hot sides of the TE modules according to their location along the length of the heat exchanger, a varying number of alternating layers of copper foil and grafoil were inserted between the heat exchanger and the TE modules. Because there was an excess of heat available from our test vehicle, we could create a larger thermal resistance at the hotter end of the heat exchanger and systematically reduce that thermal resistance for each successive row of modules. We expected that this would help to make the actual temperature profile of the TE modules much more uniform. In Fig. 23, the positions (i.e., rows) of skutterudite TE modules labeled 1 through 4 correspond to the schematic drawings of the layered thermal interfaces, shown in Fig. 26, where successively more layers were inserted in positions closer to the hotter end of the heat exchanger. A similar scheme was implemented for the Bi-Te TE module section of TEG#3 (i.e., for rows 5 – 7). All of the TE modules were placed in contact with a folded piece of copper foil (with a thermocouple inserted in the fold for only the center string), also shown in Fig. 26. The temperature profile of the cold-side heat exchanger as measured by the inlet and outlet temperatures of coolant varied only slightly along the TEG and was approximately $90 \pm 5^\circ\text{C}$. We therefore were able to use the same standard thermal paste previously used for TEG#1 and TEG#2 for thermal interface between the cold-side heat exchanger and all the TE modules in TEG#3.

The bypass valve temperature control algorithm for over-temperature protection was modified to include maximum set point temperatures for both the skutterudite and the Bi-Te TE modules. As we will describe below, the maximum temperature achievable at the front and center position of the skutterudite section (Position 1) was limited by the highest temperature allowable for the Bi-Te module (Position 5), and this limited the total output power generated for TEG#3. Installation of TEG#3 and all vehicle testing was performed on the test track and dynamometer facilities at GM's Milford Proving Grounds. Figure 27 shows the test vehicle positioned on the dynamometer.

Figure 26. Schematic drawings of copper and grafoil layers between each skutterudite module and the hot-side heat exchanger. The Position numbers correspond to those shown in Fig. 22. Thermocouples were inserted into folded copper foils and placed in contact with the hot sides of the TE modules for measuring the TE module hot-side temperatures.

Figure 27. The Chevrolet Suburban test vehicle in the dynamometer test facility: (a) the computer monitor outside the driver's door provides the US06 test drive protocol to the driver and large light blue fans behind the monitor simulate external driving conditions; (b) the rear wheel sits on the dynamometer rolls; (c) the electronics equipment in the rear compartment of the vehicle is visible through the rear hatch along with the small dry nitrogen gas cylinder used to fill the TEG to protect the skutterudite modules from oxidation; and (d) the TEG temperature and output data display is located on the dashboard and the data acquisition computer is located in front of the passenger seat.

The temperature profile of the TE modules was measured using the seven thermocouples shown in Fig. 23. The bypass control algorithm was set to limit the maximum temperature of the Bi-Te modules (Positions 5 – 7) to 250°C, as shown by the purple curves in Fig. 28. This resulted in the maximum temperature at the front and center skutterudite module (Position 1) to be limited to ~350°C. (See the red curves in Fig. 28.) The detailed results for a portion of the test cycle are shown in Fig. 29, where the vehicle speed and TE module hot-side temperature were scaled down by a factor of 10 and plotted on the same axis as the bypass valve position (% open) and TEG Output Power (W). The purple curve in Fig. 29 shows the operation of the bypass valve as it opened to allow more exhaust gas to enter the TEG as the vehicle speed fell off after about 8 minutes into the drive cycle. This resulted in a smoothing out of the temperature variation experienced by the TE modules and a smoothing out of the TEG output voltage, current, and electrical power, also shown in Fig. 29. The total output power, however, is only slightly above 25 W, and this low value is due in part to the low-temperature set point (250°C) used to protect the Bi-Te modules from overheating causes the temperature of the skutterudites to be too low (350°C) for their optimum performance, which would occur at temperatures of about 550°C.

Figure 28. Temperature as a function of time for two sequential US06 drive cycle tests: 0 – 10 min, and 11.5 – 21.5 min. The bypass control algorithm limited the Bi-Te TE module temperature to 250°C (Positions 5 – 7, purple curves). This resulted in the skutterudite module temperature to be limited to about 350°C (Positions 1 – 4, red curves), well below the temperature for optimum skutterudite performance. The exhaust gas (blue) temperature, coolant (green) temperature, and the vehicle speed (black) are also shown.

Figure 29. TEG#3 results between 5 and 9 minutes during a US06 drive cycle. Vehicle speed (black) and TE hot side temperature (red) were scaled down by a factor of 10 and plotted together with the bypass valve position (purple) and the TEG output power (brown). Vehicle speed at 5 min was ~118 kph and the TE hot side temperature was ~357°C.

The configuration of the TE modules allowed three strings of 14 series-connected TE modules to be connected in parallel outside of the TEG case. Each string consisted of 8 skutterudite and 6 Bi-Te modules connected in series. Further analysis of the performance of TE modules in our TEG#3 design was performed by averaging the temperature data for the US06 test cycle to determine the effective ΔT at each TE module and the estimated voltage, current, and power output for each module. The temperature data for the hot side of the TE module were averaged for the drive time between 5 and 9 minutes, and the results are shown in Fig. 30. From this result, ΔT was determined by subtracting the coolant temperature profile that was interpolated from the inlet and outlet temperatures for each TE module position.

These results clearly show a successful demonstration of a skutterudite-based TEG installed and tested on a production vehicle in test track driving and dynamometer drive cycle runs. The data analysis indicates that the very low power output under these test conditions arise because of the low operating temperatures necessary to protect the lower temperature Bi-Te TE modules. In addition, the temperature

profile along the length of the TE is sufficiently non-uniform so that, even if we were able to get the front row of skutterudites up to their optimum operating temperature, the subsequent rows would be significantly lower in temperature resulting in unacceptably low performance.

Figure 30. Averaged TEG#3 temperature, voltage, and power versus TE module position using the US06 test data between 5 and 9 minutes (Fig. 28). The bypass control algorithm limited the hot-side temperature (red) of the Bi-Te TE modules (Positions 5 – 7) to 250°C which resulted in the skutterudite module temperatures (Positions 1 – 4) being limited to ~375°C, well below the ~550°C optimum temperature for skutterudite performance. The exhaust gas (blue) and coolant (dark green) temperatures are interpolated from measured temperatures at the inlet and outlet, and the ΔT (purple) is the difference between the red and green curves. The estimated individual TE module output voltage (light green) and power (brown) are based on measured performance data corresponding to these operating conditions of TEG#3.

We performed a more detailed analysis based on the temperature profile measurements and the expected TE module performance based on the specifications to ascertain what the performance should be with additional improvements relating to optimizing the temperature profile. The measured temperature profile of the hot side of the TE modules is shown in Fig. 30 along with the temperatures of the exhaust

gas and coolant based on the relatively steady state portion of the US06 drive cycle between 5 and 9 minutes. This is the same time interval plotted in Fig. 29. The resulting averages are plotted as a function of position along the TEG in Fig. 30, and the results indicated that the effect of the thermal interface variation describe above has only a small effect on the temperature profile and that considerable temperature variation is still experienced by the TE modules. Based on the estimated hot- and cold-side temperatures at each position, the performance of each TE module was estimated and plotted in Fig. 29. Because all of the TE modules represented in Fig. 29 were connected in series, they all had the same current, but the module voltage and power generated by each module vary according to the operating conditions at each position. Based on these results, we see that increasing the temperature by allowing more exhaust gas to flow (i.e., adjusting the set point of the Bi-Te module at Position 5 to rise to about 300°C) will give more electrical output.

As we continued testing TEG#3, it became apparent that each 14-member series string performed somewhat differently, putting out different voltages and currents under the same operating conditions. This situation, unfortunately, also contributed to a lower power output than expected for all 42 modules connected in series. All 42 TE modules connected in series, however, resulted in too large a voltage for our dc-to-dc converter at optimum operating temperatures, and it was therefore necessary to either use parallel connections or to test each 14 TE module string separately. In the latter test, we confirmed that the power output for the individual strings was actually much larger compared to the power delivered in the parallel configuration. Fig. 31 shows data collected for one of the 14-member series connected strings of TE modules when the measured temperature of the Bi-Te modules was allowed to rise to about 310°C and the skutterudite TE module temperature rose to about 415°C. The 19 W of TEG power for this one string of TE modules (4 skutterudite and 3 Bi-Te) was 1/3 of the total TEG power available, and hence gave a total TEG power of 57 W. This corresponded to an average power output of 1.6 W for the skutterudite modules and about 1.05 W for the Bi-Te modules.

Preliminary performance testing of one of the 32-uncouple skutterudite TE modules used for TEG#3 was performed in collaboration with Dr. Jan König at the Fraunhofer IPM in Freiburg, Germany. The power output, shown in Fig. 32, was determined using a load resistance matching technique to map out the maximum power output as a function of temperature gradient, ΔT . Our results show that the performance of the skutterudite modules was quite good and comparable to PbTe TE modules in the same temperature range. Moreover, these results show that under better optimized operating conditions, i.e., a hot-side temperature of 600°C and a cold-side temperature of 100°C in TEG#3, the skutterudite modules would deliver about 8 W. The specifications for Bi-Te modules indicate that they can generate about 2.5 W per module in optimized conditions at 250°C. Under these conditions, TEG#3 would generate about 235 W.

Figure 31. Temperatures (blue, red, orange, and purple), vehicle speed (black), bypass valve position (green), and TEG#3 output power (brown) for a steady state portion of a drive test of the demonstration vehicle with TEG#3 installed. Only one of the three parallel strings of TE modules (8 skutterudite modules and 6 Bi-Te modules) were measured in this test, which generated a maximum of 19 Watts. After about 3.3 minutes, the engine speed was reduced causing the exhaust temperature to fall. To compensate for this, the bypass valve was further opened to allow more exhaust gas to flow through the TEG and help to maintain steady temperatures in the TEG.

Figure 32. Skutterudite TE module output power with results for PbTe shown for comparison.

In the module performance testing, however, the temperature was measured using thermocouples imbedded in blocks located above and below the TE module. Consequently, these measured temperatures were not the actual temperatures of the hot and cold side of the module itself, but somewhat higher and lower, respectively. Using the open circuit voltage relation for the Seebeck coefficient of the skutterudite TE materials used in the module, we deduced a temperature difference of nearly 100°C between the temperature measured and the actual temperature of the thermoelectric legs at the hot-side junction, as shown in Fig. 33. By correcting the measured output power for the skutterudite TE module, we expect that under optimum operating conditions, we would get as much as 14 W, as shown in Fig. 34. Thus, the actual power output performance for our TEG#3 with better optimized ΔT s is expected to be as high as 385 W.

Figure 33. Measured open circuit voltage V_{oc} for skutterudite modules (filled circles). The predicted V_{oc} (unfilled circles) was calculated using measured Seebeck coefficients of the skutterudite legs. The red arrow shows that there is about a 100 K drop in temperature from the hot-side heat exchanger to the skutterudite legs at operating temperatures ($T_h \sim 555^\circ\text{C}$, $T_c \sim 80^\circ\text{C}$). In other words, the measured $V_{oc} \sim 2.6$ V at the measured $\Delta T \sim 475^\circ\text{C}$ is consistent with an actual $\Delta T \sim 375^\circ\text{C}$.

The intrinsic properties of the skutterudites synthesized for the TE modules installed in TEG#3 were considerably lower than what is typically obtained in carefully optimized laboratory samples, namely $ZT \sim 1$ for n-type and $ZT \sim 0.6$ for p-type skutterudite leg material at 750 K. With further material development and property optimization, we expect that the power output performance of skutterudite modules to further increase by 10%. This means that ultimately TEG#3 would be able to generate about 425 W average power during the 5 – 9 minute portion of the US06 drive cycle. This is considerably

larger than predicted based on our thermal modeling at the beginning of this project, which was about 350 W needed for a 3% FE improvement for the Chevrolet Suburban. Overall, however, the total power output of TEG#3 would likely be reduced significantly because of the large thermal gradient along the length of the heat exchanger in spite of our attempt to introduce varying amounts of thermal interface resistance to compensate. This means that in addition to improving thermal contacts/interfaces and skutterudite material transport properties as discussed above, any future heat exchanger design must mitigate the non-uniform temperature profile that typically develops along exhaust gas flow direction in the heat exchanger.

Figure 34. Uncorrected (filled circles) and corrected (unfilled circles) output power for skutterudite TE modules. The correction is based on a calculated ΔT drop between the hot-side heat exchanger and the skutterudite legs by comparing the intrinsic Seebeck coefficient of the skutterudite material and the measured TE module open circuit voltage, as described in Fig. 33. By extrapolating the corrected power output to the operating conditions ($\Delta T \sim 475^\circ\text{C}$, $T_h \sim 555^\circ\text{C}$, $T_c \sim 80^\circ\text{C}$), we expect an output power of up to 14 W per skutterudite TE module.