High-performance nanostructured thermoelectric generators for micro combined heat and power systems

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Abstract

Micro combined heat and power (micro-CHP) systems are promising pathways to increase power generation efficiencies. Here a new class of micro-CHP system without moving parts is experimentally demonstrated by integrating high-temperature thermoelectric generators (TEGs) and residential gas-fired boilers, thus enabling wide applications. The TEGs fabricated using high-efficiency nanostructured bulk half-Heusler alloys generate ultrahigh power density of 2.1 W/cm² with 5.3% electrical efficiency under 500 °C temperature differences between the hot- and cold- side. The TEG system harnesses the untapped exergy between the combustion gas and water, and converts thermal energy into electric power with 4% heat-to-electricity efficiency based on the total heat input into the TEGs. The high-performance TEGs open lots of opportunities to transform power generation technologies and improve energy efficiency.

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1. Introduction

In recent years, micro combined heat and power (micro-CHP) systems have been of increasing interests as they offer high-efficiency and off-grid energy solutions for residential applications [1]. There are several reasons for the growing interests in micro-CHP systems. First, micro-CHP systems increase the overall fuel efficiency due to the local production and usage of heat and power at a common location, which not only avert energy losses due to transportation, but also lead to effective usage of waste heat accompanied with power generation processes. Second, micro-CHP systems can supplement the intermittence of most renewable energy systems [2], and increase energy security for areas vulnerable to power outage. Third, micro-CHP systems are attractive due to their environmental merits via the reduction of CO₂ emissions [3]. Currently, there are a few micro-CHP technologies, including fuel cells, Stirling engines, and internal combustion engines, etc. Despite the high electrical efficiency of 20-40%, the above technologies are facing challenges such as high cost or maintenance issues associated with moving parts, which limit their applications [4, 5].

Thermoelectric generators (TEGs) present a new opportunity for high-performance and low-cost micro-CHP systems. Based on the Seebeck effect, TEGs enable the direct conversion of heat into electricity without moving parts [6]. The TEG based micro-CHP system is advantageous due to its completely solid-state nature, which enables reliable, cost-effective, and quiet operation that is unattainable from most CHP technologies. The TEG heat-to-electricity conversion efficiency η_{te} can be approximated as [6],

$$\eta_{te} = \frac{T_h - T_c}{T_h} \frac{\sqrt{1 + (ZT)_M} - 1}{\sqrt{1 + (ZT)_M} + \frac{T_c}{T_h}} \quad (1)$$

where T_h and T_c are the temperatures on the hot side and cold side of thermoelectric devices respectively, and $(ZT)_M$ is the mean thermoelectric material figure of merit between T_h and T_c .

The ZT is a dimensionless number defined as $ZT = (\alpha^2 \sigma / k) T$, where α , σ and k are the Seebeck coefficient, electrical conductivity, thermal conductivity, respectively, and T is the absolute temperature [6, 7].

Besides η_{te} , the TEG electrical power density is another important criterion that directly determines the power unit cost for TEG systems [8]. Both the TEG η_{te} and power densities can be increased by improving the material ZT or increasing the temperature difference ΔT between the TEG hot- and cold- side surfaces. Since the late 1990s, the ZT of thermoelectric materials has been improved significantly through several approaches, including low-dimensional materials such as superlattice thin films, nanocrystalline structures, and nanocomposites [9-14]. Despite significant ZT increases in nanostructured materials, there have been numerous challenges in using these materials for high-temperature TEGs due to their poor thermal stability and low mechanical strength at high temperatures, as well as difficulties in making reliable metal contacts with low contact resistances [15, 16]. Several theoretical studies have identified TEGs as a promising approach for CHP applications [17-19]. However, there have been few experimental studies on TEGs for micro-CHP systems, and the majority of them were using bismuth telluride based TEGs that can only operate under hot-side temperature $< 200^\circ\text{C}$ with very low efficiency and power density [20-25].

Fig. 1. Schematic design of a micro-CHP system, using nanostructured bulk TEGs sandwiched between a combustion gas heat exchanger and the boiler water tubes, for maximum temperature gradient.

Here we report a micro-CHP system using nanostructured bulk high-temperature TEGs integrated in a gas-fired combination boiler that provides both space heating and domestic hot water (shown in Fig. 1). The TEG produces an ultra-high electrical power density greater than $2\text{ W}\cdot\text{cm}^{-2}$ under the ΔT of $500\text{ }^{\circ}\text{C}$. In addition, it should be pointed out that the TEG is implemented directly in the ultrahigh-temperature combustion gas close to the boiler burner, and thus harnesses the wasted exergy in the boiler, which is essentially different from the conventional method of waste heat recovery involving the TEG further down the exhaust stream.

2. Fabrication of high-power-density TEGs

The TEG is fabricated using the nanostructured bulk half-Heusler alloys with a peak ZT of 1.0 at $500\text{ }^{\circ}\text{C}$ (n-type) and 0.9 at $700\text{ }^{\circ}\text{C}$ (p-type), which are 25% and 80% higher than their non-nanostructured bulk counterparts due to the large reduction in thermal conductivity as a result of nanostructuring [26-30]. In addition, the nanostructured half-Heuslers have comparable

thermal stability and improved mechanical strength compared to their bulk counterparts in the operating temperature range up to 600 °C [31].

The bonding between the thermoelectric materials and metal conductors is the most important process in TEG fabrication because of the requirement for high bonding strength and low electric contact resistances. Here the n-type and p-type half-Heusler elements ($1.8 \times 1.8 \times 2 \text{ mm}^3$) were directly brazed to a thin copper plate of 0.3 mm thickness at 825 °C in vacuum using the silver and copper based brazing alloy (Incusil®-ABA™), which enables the TEG to operate reliably at hot-side temperatures up to 600 °C. This simplified metal contact processing yields not only ultra low contact resistivity of $\sim 1 \text{ } \mu\Omega \cdot \text{cm}^2$ but also very high bonding strength of 40 MPa [32]. The thermoelectric device consisting of 60 n- and 60 p- uncouples was sealed in a stainless-steel enclosure in a reduced argon environment to protect the device from oxidation. In order to improve thermal contact and relieve thermal stresses under device operation, a 0.3 mm-thick graphite foil was inserted between the enclosure and the device to provide a flexible thermal interface.

Fig. 2. (a) TEG power density vs. electric current and (b) TEG efficiency measured at various

hot-side temperatures when the cold-side temperature is maintained at 100 °C.

A customized testing system was built to measure the TEG power output and efficiency in vacuum under various ΔT . A cartridge heater embedded inside a copper block and a cold plate with circulating chilled water were used to control the T_h and T_c of the TEG respectively. A copper block of known thermal resistance was mounted between the TEG cold side and the cold plate in order to determine the heat flow rate Q through the TEG. Several thermocouples were instrumented on the TEG hot- and cold- side surfaces and inside the copper blocks in order to determine T_h , T_c and Q . A radiation shield was implemented surrounding the heater to minimize radiation heat losses, and convection losses are negligible in a high-vacuum (10^{-5} Torr) testing environment. Graphite foils were inserted between the heater, TEG and cold plate interfaces with a clamping pressure of 0.55 MPa applied during testing in order to minimize the thermal contact resistance across the TEG internal and external interfaces.

Fig. 2 (a) shows the TEG electric power density measured as a function of electric current at various hot-side temperatures when the cold side is maintained at 100 °C. The TEG generates the peak power density of $2.1 \text{ W}\cdot\text{cm}^{-2}$ at the ΔT of 500 °C based on the TEG hot side area ($2.7\times 2.7 \text{ cm}^2$), and the corresponding power density reaches $3.9 \text{ W}\cdot\text{cm}^{-2}$ calculated using the total area of the thermoelectric elements (3.89 cm^2), both of which are significantly higher than the previously reported best value under comparable operating conditions [33, 34]. The high power densities significantly reduce implementation areas and the unit cost of TEGs, which make them very attractive for high-temperature power generations where high heat flux is usually obtainable. Fig. 2 (b) shows the experimentally measured TEG efficiency as a function of the T_h when the T_c is maintained at 100 °C. The TEG efficiency reaches 5.3% at the T_h of 600 °C. The measured TEG efficiency is still 40% lower than the theoretical efficiency predicted

using Equation (1) mainly due to parasitic losses such as the thermal contact resistances between the TEG enclosure and the TE device and the heat losses from TEG hot side to cold side. The TEG efficiency could be further increased by reducing all the parasitic losses and using segmented or cascaded TEG design with increased overall ZT [36].

The TEG power density shows less than 2.5% degradation after 500 thermal cycles between 600 °C and 100 °C on TEG hot side, indicating excellent durability of both the thermoelectric materials and the bonding with the metal electrodes despite of high thermal stresses under large ΔT .

3. A new class of micro-CHP system

To develop a micro-CHP system, the high-temperature TEGs were integrated into the heat exchanger of a residential gas-fired condensing boiler of 60 kW maximum heat output capacity and 95% thermal efficiency based on the higher calorific value of the combustion fuel energy. The design of the micro-CHP system is shown in Fig. 1. As a proof of concept, 8 TEGs were installed on the two water-tube walls in the boiler combustion chamber so that the TEGs can receive the highest combustion gas temperature and the maximum heat flux (shown in Fig. 3). The TEG cold-side surfaces were placed in contact with the two water-tube walls in order to transfer the unused heat into the boiler water loop and maintain the TEG cold-side temperature.

Fig. 3. Pictures showing the TEG assembly process in the boiler system (a) Four TEGs assembled in series configuration (b) Eight TEGs assembled in the boiler primary heat exchanger (c) TEGs fully integrated in the boiler system during testing.

In addition to the TEGs discussed above, the heat exchanger is another important component that effectively increases the heat transfer rate from the combustion gas to the TEGs and thus determines the TEG hot-side temperature. As shown in Fig. 3 (b), the heat exchanger used in this work consists of an array of flat-plate fins fabricated using nickel alloy (nickel 201) due to its high service temperature, high oxidation resistance, and relatively high thermal conductivity, which meet all the requirements of this research. In order to obtain 600 °C on the TEG hot side, the heat exchangers fabricated using 0.5 mm fin thickness, 25% fin packing fraction and 40 mm fin height were attached to the TEG hot-side surfaces, between which a thin graphite foil was inserted as a thermal interface material. A clamping pressure of 0.55 MPa was

further applied between the heat exchanger and the water-tube walls in order to improve the thermal contact conductance of the TEG external and internal interfaces. The micro-CHP system was instrumented with thermocouples to measure the temperatures of the TEG hot- and cold-side surfaces. In addition, thermocouples and flow meters were used to measure the temperatures and mass flow rate of the water flowing across the TEG cold side in order to determine the TEG heat output.

Fig. 4. The electric power output and voltage of the micro-CHP system vs. the electric current tested under the medium boiler heating power.

The power output and efficiency of the 8 TEGs connected in series configuration was tested during boiler operation at steady-state conditions. The average T_h and T_c of the 8 TEGs reach 613 °C and 113 °C respectively at the medium boiler operation condition that corresponds to 30 kW of total heat output. The water temperature increase is 3.3 °C between the inlet and outlet of the TEG with a mass flow rate of 0.166 kg/s. As shown in Fig. 4, the maximum TEG power P of 94.5 W appears at an optimal electrical current of 3.5 A when the external load matches with the TEG internal resistance. The TEG system electrical efficiency η is estimated to

be 4% using the equation $\eta = P/(P+Q_{out})$, where the heat output Q_{out} on the TEG cold side is 2280 W, determined by the temperature increase of the water flowing through the TEG cold side. The TEG system efficiency tested in the boiler is 24.5% lower than the efficiency of a single TEG tested using the cartridge heater due to heat losses from the hot-side heat exchanger to the water and the temperature drop between the heat exchanger base and the TEG hot side surfaces.

The global electrical efficiency for this boiler-TEG prototype is 0.32% based on the total boiler heat input from the burner. It should be noted that the 8 TEGs implemented in this work occupied less than 10% of the total available TEG installation space inside the boiler heat exchanger, indicating significant room to scale up the electric power output and increase the global electric efficiency. To further increase the electricity to heat ratio, the TEGs can be fabricated using materials of increased ZT with segmented/cascaded designs that combine two or more thermoelectric materials operating with the peak ZT at their respective operating temperatures. In addition, the high-temperature TEGs can be used as a topping cycle to integrate with other energy converters (e.g. organic ranking cycle) as the bottoming cycle in order to fully extract the exergy in the boilers.

4. Conclusions

In conclusion, a high-performance TEG fabricated using nanostructured bulk half-Heusler alloys produces an ultrahigh power density above $2 \text{ W}\cdot\text{cm}^{-2}$ with 5.3% efficiency. A micro-CHP system of 94.5 W electrical power output is experimentally demonstrated by integrating 8 TEGs into the combustion region of a residential boiler. The electrical efficiency of the boiler-TEG system is 4% based on the heat input into the TEGs, and 0.32% based on the total heat input from the burner. The efficiency of the micro-CHP system could be further improved by integrating the high-temperature TEGs as the topping cycle and other medium-to-low

188 temperature power generation technologies as the bottoming cycle. The high-performance TEGs
189 open up many new opportunities for power generation and waste heat recovery applications.

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288

Nanostructured Thermoelectric Generator