Reproducibility and Reliability in Manufacturing New High-Temperature Thermoelectric Modules

Karina Tarantik, Martin Kluge(✉), Kilian Bartholomé, Eugen Geczi, Uwe Vetter, Mark Vergez, and Jan König

Fraunhofer-Institut für Physikalische Messtechnik IPM, Heidenhofstraße 8, 79110 Freiburg, Germany
martin.kluge@ipm.fraunhofer.de

1 The Basics of Thermoelectric Waste Heat Recovery

1.1 Introduction

What is thermoelectrics? [1] - Thermoelectrics refers to the direct conversion of an electrical current flow into a heat flow as well as the heat flow into a current flow – thermoelectrics works in both ways.

The basic principle was already recognized by Thomas Johann Seebeck in 1821. He observed that a compass needle near two different, interconnected metal wires is deflected when the temperatures at the junctions differ, whereby the degree of deflection is proportional to the temperature difference. This is due to an electrical field that is created by the temperature gradient on the conductors.

In 1834, the French scientist Jean Peltier discovered that this effect can be reversed and used as a heat pump: if a current is connected to the interconnected conductors, a temperature gradient is created at the junctions. Thermal energy is transported from one junction to the other. The so-called Peltier effect can be used for heating or cooling. The maximum possible yield when heat is thermoelectrically converted into electrical energy is physically limited by the efficiency of the Carnot cycle process.

1.1.1 Figure of Merit

In 1909 and 1911, Edmund Altenkirch introduced a constant property model to derive the maximum efficiency of a thermoelectric (TE) generator as well as the performance of a cooler, when the design and operating conditions where fully optimized [2]. This relationship, later developed into the 'figure of merit' ZT, and revealed that good thermoelectric materials should possess large Seebeck coefficients (S), high electrical conductivity (σ) and low thermal conductivity (λ). The mathematical relation is shown in Eq. 1. Along with reproducibility and reliability, ZT has been the main driver of thermoelectric materials development in recent history.

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$$ZT = \frac{S^2\sigma}{\lambda}T \tag{1}$$

Various TE material classes with high ZT values are known and recognised as suitable for the conversion of waste heat into electrical energy. The maximum ZT value (ZT_{max}) is attained at different temperatures depending on the material class and the exact composition. However, the average ZT value (ZT_{av}) across a specific temperature range is required to determine the material efficiency for a certain temperature combination (Eq. 2),

$$\eta = \frac{\sqrt{1+ZT_{av}} - 1}{\sqrt{1+ZT_{av}} + \frac{T_c}{T_h}} \cdot \eta_{Carnot} \quad \text{with} \quad \eta_{Carnot} = \frac{T_h - T_c}{T_h} \tag{2}$$

with T_c: cold side temperature (heat sink) and T_h: hot side temperature (heat source).

Figure 1 shows the conversion efficiency of TE converters, as derived from their material property ZT versus the Carnot efficiency. State-of-art converters exhibit a peak ZT value of approximately 0.8–1.0 and reach efficiencies in the range from 5–10%.

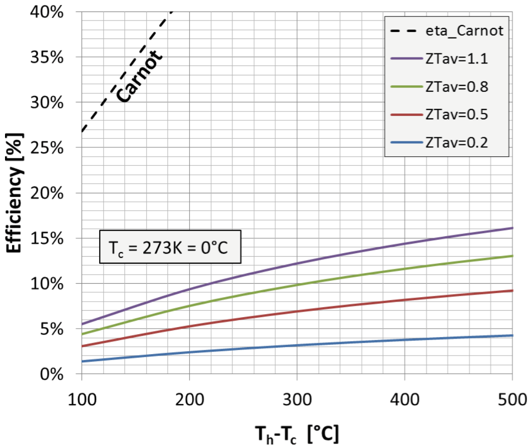

Fig. 1. Conversion efficiency at certain ZT values vs. Carnot efficiency

1.2 System Design Aspects

In a system design, the TE material is typically applied in the form of TE modules which consists of n- and p-type TE materials (legs) that are connected thermally parallel and electrically in series (see Fig. 2). Waste heat is converted by means of a heat flow through the TE module which generates an electrical current in the module. The electrical connection between the TE legs is realized in most cases by bonding the legs onto a contact material, usually a metallic strip. The contact resistance between the TE legs and the contact material should be very low in order to reduce losses caused by

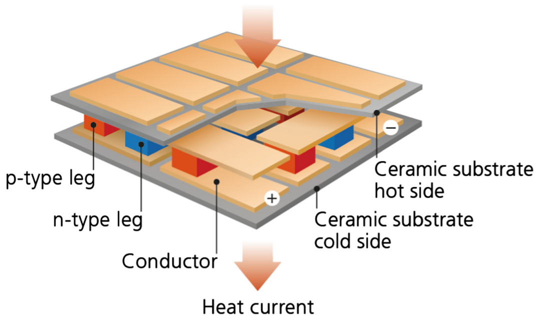

Fig. 2. Schematic view of a thermoelectric module.

Joule heating. The backbones of the TE module are two thin and thermally high conductive but electrically isolating substrates, most commonly ceramic plates. The numbers and dimensions of the TE legs differ according to the intended application of the module.

The thermoelectric modules are placed between a hot source, e.g. an exhaust gas heat exchanger, and a cold sink, e.g. a fluid cooler/heat exchanger. In combination this arrangement forms a thermoelectric generator (TEG). When designing the TEG the thermal resistance of the modules as well as the electrical load resistance of the system have to be carefully balanced in order to obtain the maximum power output [3, 4]. Due to design fundamentals, the system efficiency is typically less than 50% of the conversion efficiency of the thermoelectric material.

2 High Temperature Module Manufacturing

2.1 The Half-Heusler Alloys

Among the numerous materials studied at the Fraunhofer IPM (skutterudites, silicides, chalcogenides, …), the half-Heusler alloys have emerged to be the most reliable and most durable material class for high temperature TE applications. It allows for a usage range of up to 600 °C for its active material temperature. This high working temperature yields high conversion efficiencies in applications that are currently not accessible with commercial Bi_2Te_3 materials due to thermal limitations at above 250 °C.

After a successful scale up of powder and bulk material production in the public funded project “thermoHEUSLER” [5, 6], the developments at Fraunhofer IPM continued towards a semi-automated module production line.

Figure 3 shows the half-Heusler material properties that have been obtained by the developing partners Isabellenhütte-Heusler GmbH & Co.KG and Vacuumschmelze GmbH & Co.KG.

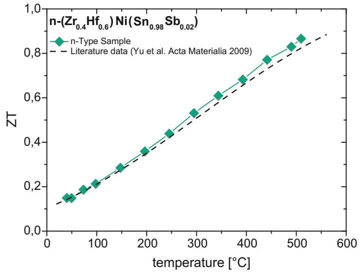

Fig. 3. Material properties obtained by Isabellenhütte Heusler GmbH & Co.KG (n-type, left) and Vacuumschmelze GmbH & Co.KG. (p-type, right) [7].

2.2 Module Manufacturing Process

The production of TE modules can be described with the following process steps (Fig. 4):

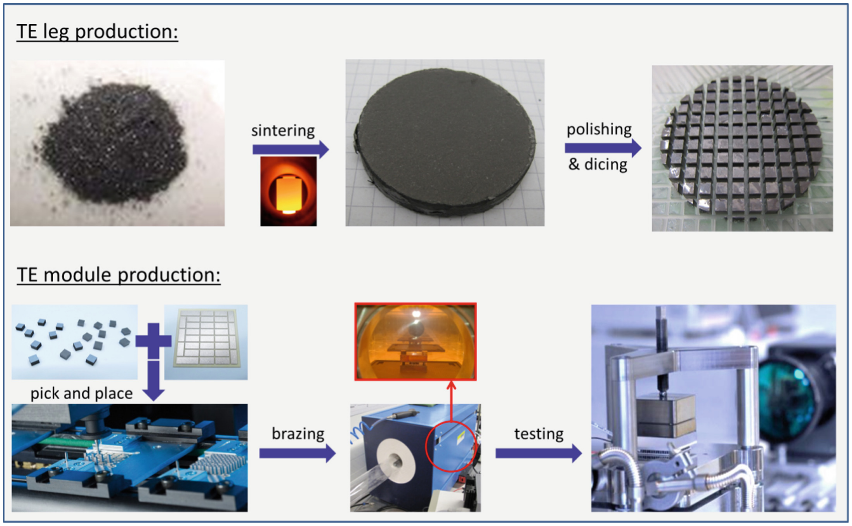

Fig. 4. TE leg and module production steps at Fraunhofer IPM.

TE leg production

- Sintering of powders: N- and p-type semiconductor pucks are compacted and sintered from powder to optimize the TE properties.
- Polishing: During the polishing step the TE leg height and surface quality are adjusted.
- Dicing: The legs are cut into their final shape.

TE module assembly

- Pick and place: N- and p-type legs are arranged on a metallized substrate ceramic (usually Al_2O_3).
- Brazing: The substrate and legs are joined using high temperature brazing process.
- Wiring/interconnection: Lead wires are attached to the module.
- Final Quality Check: Key characteristics are validated.

A cost effective and reliable module production process is essential for the industrialization of thermoelectric materials in waste heat recovery.

2.3 Recent Breakthrough

In July 2016 a new semi-automated production line has been put into operation at Fraunhofer IPM. With the new pilot-line, it is now possible to reduce the module production costs by a factor of 10. This allows moving a significant step closer towards the industrialization of the technology and opening the possibility of small series production.

2.4 Module Performance

The current top-selling module design exhibits a thermal contact area of 16×16 mm^2 and involves 7 p-n couples (Fig. 5). With a demonstrated power density of up to 1.1 W/cm^2, the module has achieved conversion efficiencies of up to 5.2% (both at $\Delta T = 530$ K, $T_{cold} = 20$ °C). By scaling the leg geometry the module can be individually matched to meet the application.

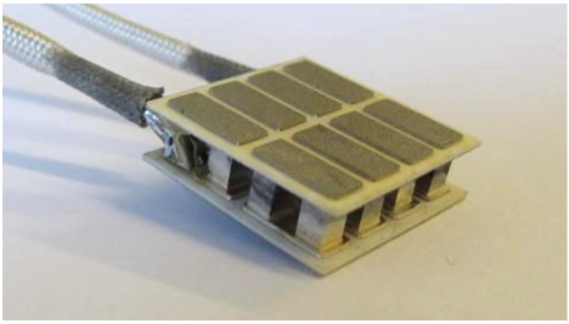

Fig. 5. Half-Heusler module made by Fraunhofer IPM.

2.5 Manufacturing Reproducibility

During module production, manufacturing reproducibility is ensured by both in-line and offline measurements. Identified key characteristic parameters which are checked are the module height and the module internal resistance at room temperature. Reasonable reproducibility values have been achieved for both:

$$\begin{array}{lll} \text{Module height deviation :} & <1\% & \text{(out of 160 samples)} \\ \text{Module internal resistance deviation :} & <6\% & \text{(out of 160 samples)} \end{array}$$

2.6 Reliability

Thermal cycling tests performed by Faurecia Emission Control Technologies, Germany GmbH in Augsburg have confirmed that the TE modules are durable enough to withstand 1,000 thermal shock cycles between 20 °C and 550 °C exhaust gas temperatures, even when being operated in air. During and after the test no significant degradation was observed. Figure 6 shows the applied test cycle and the test fixture. During the test the module's cold side temperature was maintained constant.

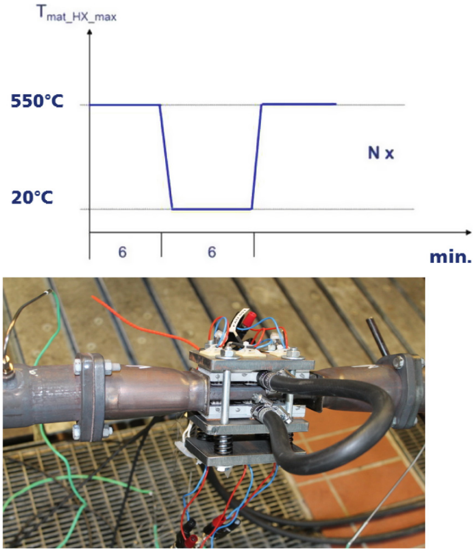

Fig. 6. Thermal cycle and test fixture.

2.7 Outlook and Future Challenges

The future of high temperature modules is tied to the identification of the right application. Today three major scenarios are considered: Automotive exhaust heat recovery, industrial waste heat recovery and combined heat and power systems (both industrial and residential). All applications inherit their need of a sufficient amount of prototypes and demonstrators to develop the system architecture and to assess economical and technical feasibility.

Despite the application, several challenges remain that need to be addressed by various means. Some of them and possible solution strategies are listed

Achieve a module cost reduction by …

- Fully automated, lean production with new and innovative production technologies
- Optimization of material usage (TE module downsizing), new and improved module designs
- Substitution or reduction of rare elements (like Hafnium)

Achieve a module performance improvement by …

- Continuous design improvements
- Advancements in TE material development

Achieve a module reliability improvement by …

- Advanced design methodologies and new module designs
- Process optimization and advanced in-line inspection technologies
- Extensive testing of modules designed for serial manufacturing

2.8 Summary

With its new semi-automated production line and advanced module manufacturing technology, Fraunhofer IPM has demonstrated that cost reduction, reproducibility and reliability are possible for high-temperature thermoelectric modules. We are ready to supply custom designed TE modules for thermoelectric generator applications ranging from automotive exhaust heat recovery to combine heat and power plants to industrial waste heat recovery.

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