CRITICAL REVIEW OF THERMOELECTRICS IN MODERN POWER GENERATION APPLICATIONS

by

Khalid M. SAQR and Mohd N. MUSA

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The thermoelectric complementary effects have been discovered in the nineteenth century. However, their role in engineering applications has been very limited until the first half of the twentieth century, the beginning of space exploration era. Radioisotope thermoelectric generators have been the actual motive for the research community to develop efficient, reliable and advanced thermoelectrics. The efficiency of thermoelectric materials has been doubled several times during the past three decades. Nevertheless, there are numerous challenges to be resolved in order to develop thermoelectric systems for our modern applications. This paper discusses the recent advances in thermoelectric power systems and sheds the light on the main problematic concerns which confront contemporary research efforts in that field.

Key words: *thermoelectrics, thermoelectric generator, waste heat recovery*

Introduction

Early history of thermoelectrics

Although the thermoelectric complementary effects were discovered in the first half of the 19th century, the first thermoelectric generator (TEG) was introduced in 1864 by Markus [1]. Subsequently Becquerel introduced his gas-fired TEG in the same year utilizing copper sulphide and German silver for the thermoelectric junctions [2]. Clamond *et al*. presented a TEG in 1879 utilizing zinc-antimony and iron for the first time. Their TEG purpose was powering electrolysis processes for metal coating [1]. Several attempts followed in the next years, until the first commercial TEG appeared in UK in the year 1925 for powering radio devices. The first commercial TEG, known as Thermattaix, used a gas burner to heat the hot junctions and ambient air for the cold junction. It is very important to note that all these early TEGs used metal based thermoelectric elements, which had quite low conversion efficiency which resulted in larger size systems.

Thermoelectric materials based on semiconductors were firstly investigated in the former USSR at the beginning of the 20th century. During the 1920s, numerous studies of mechanical, electrical, and thermal properties of crystals were carried out by Ioffe at the Physical-Technical Institute (PTI), USSR. By the years 1929-1930 he predicted a coefficient of performance value as up to 2.5-4% for semiconductor thermoelectric generator [3]. Six years later, it was shown at PTI that it was possible to create a semiconductor of both N-type and P-type by the simple change of sulphur content relatively to the stoichiometric composition for thallium sulfide [4]. Based on these findings, the solid-state physics laboratory of PTI under Y. P. Maslakovets worked in Moscow for the Intelligence Service of the General Headquarter of the Red Army to develop a special type of TEG. This TEG was intended for the electrical supply of radio station "Sever" which ensured bilateral intercommunication for the distance of 1000 km. An experimental series of TEG was produced and used in the Army in the end of the Second World War without any arguments [5].

The first commercial TEG based on semiconductor doped thermoelements was firstly produced in 1954 in the USSR. The P-type ZnSb thermoelectric elements developed by PTI were used in a ring-shaped structure to recover heat from kerosene lamp to power radio devices. This TEG became very popular at that time for its light weight and suitability for rural areas, and it extended to several parts of the world including the Middle East [5].

The 1950s have witnessed the discovery of BiTe as a base for efficient thermoelectric materials. In 1952 a thermoelectric refrigerator was fabricated with P-type elements from Bi_2Te_3. In 1956 an effective $Bi_2 (Te, Se)_3$, material for N-type leg was developed in PTI by S. S. Sinani group. In 1956 Ioffe and his coworkers generalized their observations on thermoelectric properties of solid solutions and concluded that they often could have practically the same values α and σ as basic components but significantly lower lattice thermal conductivity, *i. e.* higher *Z*. They stated that the formation of solid solutions of isomorphic thermoelectrics is a general method of increasing of thermoelectric figure of merit [6].

Thermoelectrics and space exploration era

In parallel with USSR strides in thermoelectrics research, the USA was motivating several research projects in the same field. Substantial funding from the American department of energy was directed to develop highly effective thermoelectric materials to be used in powering space exploration missions of that time. This research intensiveness was parallel to the revolutionary investigation of nuclear power sources (NPS) in the early 1950s [7].

At that time, thermoelectric energy conversion systems were characterized to have discrete potentials over other types of energy conversion systems. The key advantages of radioisotope thermoelectric generators (RTGs) are their long life, robustness, compact size, and high reliability. RTGs are able to operate continuously, and are relatively insensitive to radiation and other environmental effects. Thermoelectric converters are easily scalable, and possess a linear current-voltage curve, making power generation easy to control. They produce no noise, vibration, or torque during operation. For these reasons, US's National Aeronautics and Space Administration (NASA) used RTGs in 29 missions since 1961. Some of the most notable RPS flights are the Apollo lunar missions, the Viking Mars Landers, Pioneer 10 and 11, and the Voyager, Ulysses, Galileo, and Cassini outer planet spacecraft [8]. In fact, the simple theory of operation of RTGs is based on the decay of the radioisotope core which produces heat. This heat is transferred through thermoelectric pairs which are connected thermally in parallel and electrically in series, consequently they produce electric power.

The power generated by RTGs launched by NASA ranged from three to few hundreds of watts. Figure 1 shows the development of generated power from RTGs during the space exploration history of USA. Except for the SNAP-1OA reactor, all of the US NPS lanuched in the 1960s and the early 1970s used telluride (usually lead telluride, PbTe) thermoelectric materials to form the elements of the converter. All of these telluride-based RTGs – except the Transit satellite – operated by means of conduction between the plutonium heat source and the thermoelectric elements. Large bulk insulation was used to minimize heat losses and a cover gas was used to retard sublimation of the thermoelectric material at the hot end of the pairs [8]. The Transit

RTG operated in a vacuum using a radiant heat transfer between thermoelectric elements and the heat source. To control sublimation the Transit RTG operated at a lower hot junction temperature than did the other telluride generators. The SNAP-3B7 RTG on the Transit 4A satellite also operated under vacuum conditions to minimize conduction losses through the insulation [9, 10].

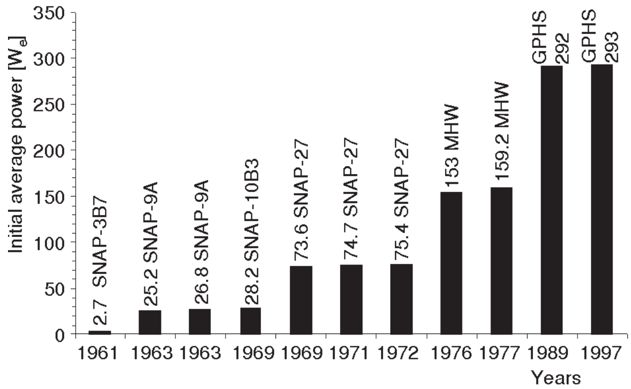

Figure 1. Development of generated power by RTGs in US space exploration missions

State-of-the-art of thermoelectric generators

Thermoelectric materials

Presently, thermoelectric materials for power generation purposes are based on PbTe at low and moderate temperature gradient applications [11, 12]. In higher temperature gradients, SiGe alloys are employed [13]. FeSi is used for high temperature applications as well, in addition, iron silicates have a very strong durability at high temperatures and mechanical stresses [14]. The advances in thermoelectric figure of merit during the last 60 years were astonishing, taking the Z value from 0.5 in late 1950s to 3.5-4 in 2007, the whole development of materials during this era is summarized in fig. 2 [14]. In the early 1990s, Skutterudites emerged as a promising class of materials for thermoelectric conversion applications in the 500 to 900 K temperature [15].

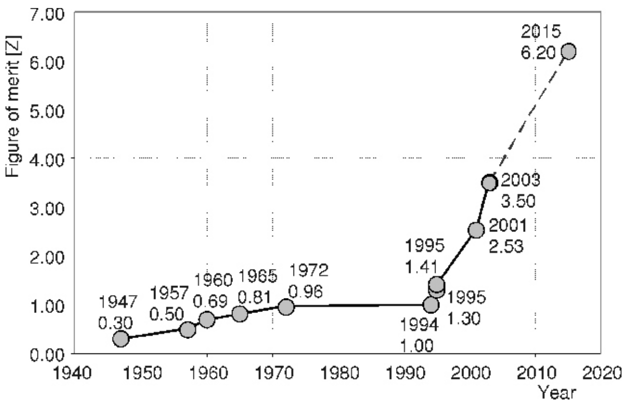

Figure 2. Development of thermoelectric figure of merit during the last 60 years

The US Department of Energy hypothesizes that the implementation of quantum-well technology in thermoelectric materials promises to increase the efficiency of thermoelectrics as energy conversion alternatives to reach 35% by the year 2030. This enormous increase in efficiency is predicted to go in parallel with the sharp fall in the price per watt for TEGs to few cents [16]. On the other hand, the revolutionary research investigating the employment of organic polymers [17] as thermoelectric materials within nano-hybrid materials opens unlimited horizons and uncountable applications of thermoelectrics as power sources in the 21st century [18, 19].

Thermoelectric generators for automotives

Internal combustion engines are well known for their low fuel conversion efficiency. Contemporary engines lose more than 60% of fuel energy as waste heat. This huge loss has been recognized as potential target for thermoelectric waste heat recovery, in order to find future solutions to reduce the running cost of passenger vehicles, and to meet the unrelenting increase in electric power demand in modern vehicle systems [20]. The first TEG for waste heat recovery in automotive has been developed in 1963 [21]. Several researches followed in the second half of the 20th century. The most remarked prototypes were developed by Porsche [22], Hi-Z [23, 24], Nissan Motors [25], and Clarkson University in collaboration with GM [26, 27]. The power produced by these generators ranged from 30 W to 1000 W from gasoline and diesel engines at different testing conditions. All of these TEGs have used exhaust gases and engine coolant as the heat source and sink, respectively. The main characteristics of these TEGs are summarized in tab. 1. Enhanced power density TEGs have been reported by the authors in several literature [28, 29]. Some of the reported work identified power density of up to 6.92 W/kg for 2.0 l gasoline engines [30].

The economic impact of implementing TEGs in today's road transport sectors is very promising, especially with the recent global economic crisis and the extensive fuel price increase. A recent study was performed on the deployment of TEG as a legally required component in the Malaysian road transport sector [31]. The authors in this study approximately determined the annual market size of automotive TEG of 242 million USD. The economic benefit comes from the ability to replace the alternator with a retrofitted TEG, giving the engine more fuel, and less load to supply.

Table 1. Characteristics of the state-of-the-art TEGs for automotive waste heat recovery

Characteristics	Hi-Z TEG	Nissan TEG	Clarkson TEG
Engine	Cummins 14 l diesel engine	3.0 l gasoline engine	8.0 l GM gasoline engine
Testing condition	1700 rpm 220.65 kW (300 hp)	60 km/h – Hill climb mode	112.65 km/h – Hill climb mode
Thermoelectric materials	72 HZ-13 BiTe modules	72 SiGe modules	16 HZ-20 BiTe modules
Thermoelectric conversion efficiency	4.5%	1~2%	2.8-2.9%
Total weight	44 kg	14.5 kg	39.1 kg
Power density	24.27 W/kg	2.45 W/kg	6.52 W/kg

Thermoelectric waste heat recovery from industrial applications

The largest part of the world's overall energy resources has been consumed as thermal energy at a reported mean efficiency of not more than 30 to 38%. After utilization the major portions of fuel energy are finally rejected to the atmosphere, rivers, or oceans as waste heat [32]. The existing technologies for efficient use of energy and recovering waste energy have almost reached their limitation, consequently the development of innovative technologies for various energy fields, such as energy conversion, energy storage, transmission, and unexploited energy utilization is urgently required. Thermoelectric power generation stands as a sole potential technology to achieve higher efficiency for industrial systems in the near future.

As an example, in Japan, the government has supported several researches to develop efficient thermoelectric technologies for industrial waste heat recovery since the early 1990s [33]. One of the most successful researches was the development of a 500 kW thermoelectric system for waste heat recovery in a pressurized water reactor (PWR) power plant [34]. Steam is continuously released from PWR steam generator blowdown system in order to keep water purity at the desired level. In the mentioned case, wasted steam of 220 °C, 97 ton per hour is released from an 1100 MW_e (3200 MW_t) PWR power plant. Thermoelectric modules were sandwiched between hot ducts, which having the steam, and cold ducts having cold water at 20 °C, as in fig. 3. The thermoelectric modules were designed and fabricated with a new technique to dramatically reduce the thermal contact resistance and cause thermal stress relaxation in the thermoelectric material. This novel technique resulted in achieving three times higher thermal efficiency for the mentioned system. The thermal transfer analysis revealed that the thermoelectric system provides AC 500 kW_e. The electric cost of 8.5 yen/kWh (0.068 USD/kWh) would be achieved in the future.

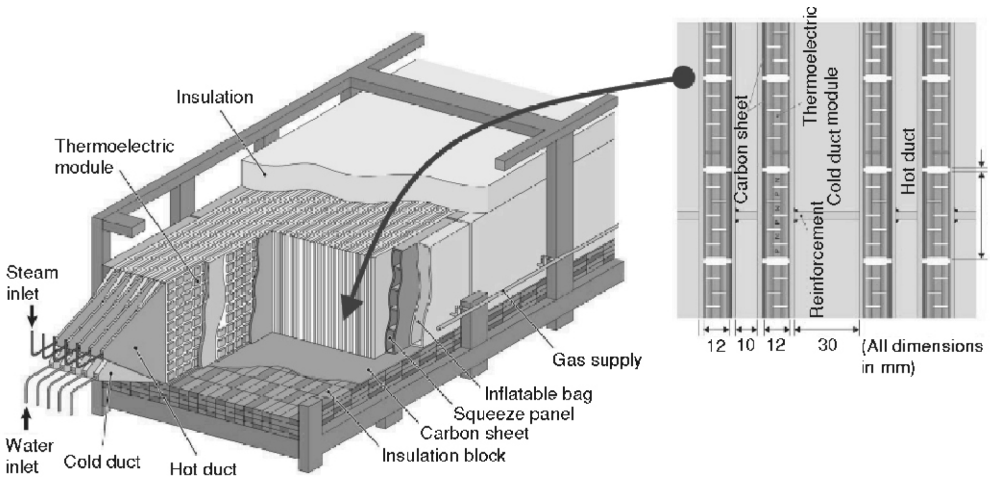

Figure 3. 500 kW TEG for waste heat recovery from PWR power plant, Japan

Another example in the oil and gas industry is the TEG developed by Hi-Z Inc. to recover waste heat from triethylene glycol dehydrators which is essentially used in remote inaccessible natural gas extraction sites [35]. The thermoelectric generator was fabricated from mild steel; it consisted of a single high temperature heat exc

hanger of rectangular cross-section, eight HZ-14 thermoelectric modules, two cold side heat exchangers, and spring-loaded clamping system to hold the cold side heat exchangers and thermoelectric modules in good thermal contact against the central hot heat exchanger. The system produces 60 W of electrical power for cathodic protection supply. However, the system was capable of producing 100 W at higher temperature difference between the hot and cold sides.

The commercial TEGs produced by Global Thermoelectrics Inc. for oil and gas industry are the most commonly used power sources for remote SCADA and emergency communications in offshore extraction fields and pipelines. The generated power of GT TEGs varies from 60 W to 6 kW. In the present time, more than 20.000 units are installed in 50 countries worldwide [36]. Basically, these TEGs are fossil fuel powered; they include a burner, combustion chamber, and a cold-side heat exchanger as a heat sink. The type of fuel is dependent on the application addressed; for example, if the TEG is installed to power the SCADA instrumentations in a natural gas pipeline, the TEG uses a bleeding sub line to produce the required heat.

Thermoelectric waste heat recovery from human body

The identification of human body heat as a target for thermoelectric waste heat recovery was parallel to the substantial spread of mobile computing, telecommunications, and entertainment in the early 1990s. However, it took several years to produce actual experiments in this critical application. The requirements of medically safe materials to be attached to the human body, light weight and flexibility are restricting the development of power systems that can recharge cell phone batteries, gaming stations, notebooks, and ipods using the human body heat.

In 2007, scientists from Spain and Germany introduced a TEG based on human heat harvesting to power wireless sensors [37]. They reported that the TEG is capable of generating 2.05 mW at 334 mV at a hot side temperature of 312 K and temperature difference of 6.71 K with an overall efficiency of 0.17%. The design included a miniaturized DC-DC voltage boosting circuit and an energy storage element. EnOcean RF sensor transmitter STM 100 and receiver RCM 120 modules have been selected as application examples to validate the TEG performance. It was found that the energy harvested from the contact with a human hand suffices to power the sensor unit and to transmit the required data every 1 second by means of the mentioned wireless communication module.

Earlier last year, scientists in Lawrence Berkeley National Laboratory and the University of California revealed some information about their research on implementing silicon nanowires in harvesting the human body heat [38]. In fact, silicon nanowires have relatively high figure of merit at room temperature, which qualifies the newly developed material to play the major role in such an application.

Opportunities and challenges

Thermoelectric materials

The prospects of thermoelectric materials to be employed in waste heat recovery have increased significantly with the advances in semiconductor fabrication technology and the emergence of novel materials. The basic challenge facing the development of new thermoelectric materials is maximizing the figure-of-merit Z. The optimization of the figure-of-merit appears to be a challenging problem since the three material properties that determine this quantity are closely connected. Therefore current researches are supposed to find ways, in principle, to optimize the figure-of-merit by suggesting materials with optimized electronic band structures and thermal transport properties.

Another important challenge is to the operating temperature of these applications. Figure 4 explains the range of operating temperatures for different prospective applications. Mod-

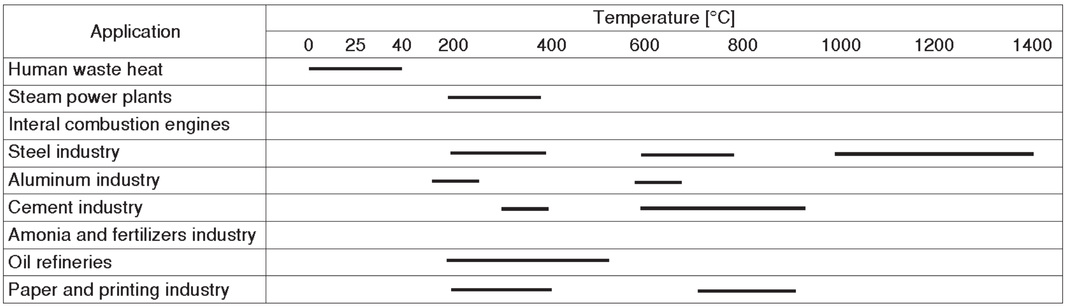

Figure 4. Range of operating temperatures for thermoelectric waste heat recovery

ern commercially available thermoelectric modules can operate only in the range of 150 ºC to 400 ºC covering a limited number of applications. New thermoelectric materials should be developed to cover lower temperature ranges for human waste heat, and higher temperature ranges to cover metal industry applications.

The third and most difficult challenge facing the development of new efficient thermoelectric materials is concerned with the module shape. In the present time, modules are produced in a rectangular shape only, which dramatically restricts the geometry of TEGs. This restriction comes from the demand of providing flat surfaces to mount the module on. For example, in order to recover waste heat from a steam pipe, a heat exchanging element should be developed in order to contact the pipe cylindrical surface – *or the hot steam* – from one side, and the module flat surface from the other side. This heat exchanging element, in many cases, reduces the thermal efficiency of the TEG significantly.

There are very few researches addressed this challenge so far. A flexible polymer based micro thermoelectric wafer was introduced by Swiss scientists in 2006 [39]. The thermoelectric wafer was fabricated by subsequent electrochemical deposition (ECD) of Cu and Ni in a 190 μm thick flexible polymer mold. At temperature difference of 0.12 K, the wafer generated 12 1.1 μW/cm^2. The other researches on fiber based thermoelectric materials are published this year by researchers from USA [40], and Portugal [41]. The main target for the both researches is clearly to develop human body waste heat recovery for powering mobile electronics.

TEG enclosing thermal systems

The general objective of thermal systems designed to contain thermoelectric materials for power generating applications is to provide efficient heat transfer characteristics. These characteristics include high thermal effectiveness, uniform temperature gradient, and high temperature difference between hot and cold surfaces of the thermoelectric modules. In order to develop such systems, modern heat transfer enhancement techniques, new materials, and innovative geometries should be employed.

As to automotive exhaust based TEG systems, the main thermal challenges have been identified to be [42]:
- heat exchanger geometry,
- heat exchanger materials,
- the installation site of the TEG, and
- the coolant system of the TEG.

The response of engine and its cooling system to the TEG should be carefully investigated to prevent any backpressure effect on the exhaust manifold causing inefficiency, and to minimize the added cooling load on the radiator [42]. The installation site should be optimized with respect to exhaust gas temperature at different engine performance conditions, in order to stabilize the generated power.

In industrial applications, the containing thermal systems should provide significantly high thermal efficiency in order to harness the major portions of waste heat. Heat sinks based on ambient air should be employed in order to minimize the TEG system complexity and maintenance demands. Furthermore, the potential of TEGs as additional power sources should be evaluated in comparison with the total power consumption of the industrial plant with respect to the timely increase in energy prices in order to recognize the economic benefit of the TEG.

For human waste heat applications, research work should focus on finding new materials that are body-friendly, and having good thermal conductivity in the same time, in order to build efficient TEG thermal containing systems. The main characteristics of the thermal systems for human heat recovery TEG are firstly to have low density, high flexibility to eliminate any motion restriction, and high durability. The potential materials to provide these characteristics are polysulfones, composite fibers, and liquid crystal polymers.

Conclusions

Thermoelectric power generation is an emerging technology, proposed as a highly alternative energy conversion system to increase the efficiency of current power producing technologies, as well as providing innovative energy solutions for present and future demands.

Research in thermoelectric power generation, especially in waste heat recovery, takes three major trends; one is the thermoelectric materials, two is the enclosing thermal systems, and three is the energy efficiency and economics.

Present research on thermoelectric materials aims to develop materials with high figure-of-merit at a broader range of operating temperatures, in order to cover more applications. The demand for flexible thermoelectric materials is persisting for the potential future application of human body waste heat recovery.

The development of highly efficient containing thermal systems is necessary to have successful TEG designs in any application. Material selection plays a major role in designing effective heat transfer systems. In addition, innovative geometry can fundamentally enhance the thermal efficiency of forced-convection heat transfer systems. TEG containing thermal systems should be medically safe, flexible, and light weight for human body waste heat recovery applications.

Accurate, up to date, and inclusive economic analysis for achievable thermoelectric savings should be carried out in order to identify the boundaries for this technology and motivate different original equipments manufacturers and markets to accept it.

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Authors' affiliation:

K. M. Saqr (**corresponding author**)
Thermofluids Laboratory E06 # 110
Department of Thermofluids, Faculty of Mechanical Engineering,
Universiti Teknologi Malaysia
81310 Skudai, Johor Bahru, Malaysia
E-mail: mmdmskhalid2@siswa.utm.my
khaledsaqr@gmail.com

M. N. Musa
Department of Thermofluids, Faculty of Mechanical Engineering,
Universiti Teknologi Malaysia
81310 Skudai, Johor Bahru
Malaysia

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