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HIGH ENERGY DENSITY MINIATURE ELECTRICAL AND THERMAL POWER SOURCE USING CATALYTIC COMBUSTION

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ABSTRACT

This paper describes the development of a thermoelectric (TE) generator with a catalytic combustor. The TE generator is composed of a thermoelectric (TE) module, a catalytic combustor and an ignition heater. The generation test was performed by attaching two TE modules to the combustor and using methanol as fuel. The conversion efficiency reached 2.39 % at an output power of 294 mW. Starting-up time was shorter than 5 sec after fuel was supplied. When restarting combustion, the generator produced power several times without damaging a catalyst.

1. INTRODUCTION

Liquid hydrocarbon fuels have more energy per volume and weight than the best existing batteries. For example, the energy density of a lithium ion battery is about 200 Wh/l and 100 Wh/kg. The energy density of gasoline is over 13 kWh/l and 15 kWh/kg. This suggests that 2-10 % conversion efficiency of these fuels offers more energy compared to the lithium ion battery. In addition, refueling is faster than recharging batteries. Used batteries are not easy to recycle, although they cause environmental problems when discarded. Also, users often feel confusion and inconvenience to dispose used batteries. Replacing batteries with reusable micro-generators and fuel cartridges made of recycled plastics will relax the environmental problems of batteries.

From the above reasons, researches to generate power from liquid hydrocarbon fuels for portable computers or mobile machines are collecting much attentions. For example, gas turbines, fuel cells, fuel processors, thermoelectric (TE) generators[1,2], thermophotovoltaic generators[3,4] and a thermoionic generator[5] are under development.

TE generation basically needs no moving parts, and the conversation efficiency of several percentages, which is enough to exceed the energy density of lithium ion batteries, is possible using available TE materials. In this study, we have developed a miniature TE generator with a catalytic combustor for applications where exhaust heat is also useful.

In our previous study, the combustor without a load (TE module) achieved self-sustaining combustion and electrical ignition using butane as fuel. When the TE modules were attached to the combustor, however, butane combustion was impossible and the combustion efficiency of hydrogen decreased. Thus, the generation test was performed by using hydrogen as fuel. The conversion efficiency reached 1.8 % at an output power of 276 mW. In this study, we tested the generator using methanol as fuel, because methanol will become a common fuel for portable fuel cells.

2. SYSTEM CONCEPT

Figure 1 shows the system diagram of the thermoelectric generator with a catalytic combustor. This system is composed of a thermoelectric (TE) module, a catalytic combustor, an ejector to supply air to the combustor and an ignition heater. Liquid gases such as butane and propane are used as fuel. The liquid gas is ejected from a nozzle in the ejector by its own vapor pressure, making supersonic jet. Air is sucked by viscous dragging and pressure drop due to the fuel jet, and supplied to the combustor with the fuel. Using this system, self-sustaining generation without a fuel/air pump becomes possible.

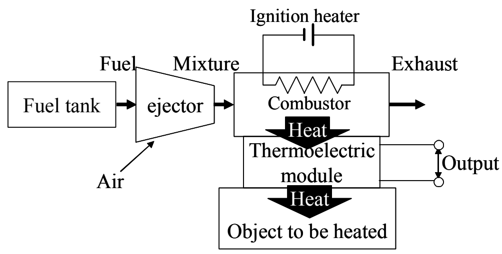

Fig 1: System diagram of the thermoelectric generator

The maximum output power of TE generation, P_{max}, is given by

$$P_{max} = \frac{ZK\Delta T^2}{4}, \quad (1)$$

where Z is figure of merit, K is total thermal conductance, and ΔT is temperature difference between a hot and cold junction. The maximum efficiency, η_{max}, is given by

$$\eta_{max} = \frac{\Delta T}{Th}\frac{\sqrt{1+Z\bar{T}}-1}{\sqrt{1+Z\bar{T}}+Tc/Th}, \quad (2)$$

where $\bar{T}$ is the average of hot and cold junction temperature, *Th* and *Tc*. Under the assumptions that Th = 150 ºC and Tc = 50 ºC and $Z = 2.7 \times 10^{-3}$ K^{-1}, the efficiency of TE generation becomes $E \approx 5$ % from Eq. (2).

If we can attain 90 % combustion efficiency (Ec = 0.9), and 30 % heat loss (Lh = 0.3), the total efficiency of the generator, *Et*, becomes

$$Et = E \times Ec \times (1 - Lh) = 3.2\ \%.$$

The total efficiency of 3.2 % corresponds to an energy density of 400 Wh/l and 3200 Wh/kg by using hydrocarbon fuels with an energy density of 13000 Wh/l and 100000 Wh/kg. This energy density is about 32 times higher than that of existing lithium ion batteries (about 100 Wh/kg) per weight and about 2 times higher (about 200 Wh/l) per volume[6].

3. STRUCTURE

Figure 2 shows the structure of the TE generator. A combustion chamber is etched in a silicon substrate, and bonded to both side of a glass substrate with a thin-film ignition heater. One side of TE elements is directly bonded on the backside of the combustion chamber where the interconnection electrodes are formed. The other side of the TE elements is bonded to the AlN bottom substrate with the interconnection electrodes and output terminals. Fuel-air mixture is supplied from a glass tube. Figure 3 shows the photograph of TE generator.

In a current prototype, the outer size of the combustor is 14 mm × 10 mm × 1.8 mm, and the volume of the combustion chamber is 8 mm × 8 mm × 0.4 mm × 2 (0.05 cm^3). The combustion chamber is coated with Pt catalyst loaded on TiO_2 support made by sol-gel method, allowing stable combustion to be maintained in the small space under the quenching distance at relatively low temperature (200-400 ºC). It should be noted that minimizing pressure loss is essential to use the ejector.

A BiTe system is used as TE material. Micro-TE generators using thin film TE materials[1,2] and a TE module using electroplated TE materials[7] have been reported, but we have selected bulk materials to obtain the possibly highest performance. The p-type material is Bi_2Te_3-Sb_2Te_3, and the n-type material is Bi_2Te_3-Bi_2Se_3. These materials have a Seebeck coefficient of 205 μV/K, a resistivity of 1.1 mΩcm and a thermal conductivity of 1.3 W/mK, that is, $Z = 2.7 \times 10^{-3}$ K^{-1} at 300 K in average.

In the current prototype, 34 couples of the TE element are arrayed in an area of 7.8 mm × 9.6 mm to achieve 1.5 V output at ΔT = 100 ºC. The size of each TE element is 0.65 mm × 0.65 mm × 2 mm t. In this case, the thermal conductance of the TE module is K = 17 mW/K. The specifications are summarized in Table 1.

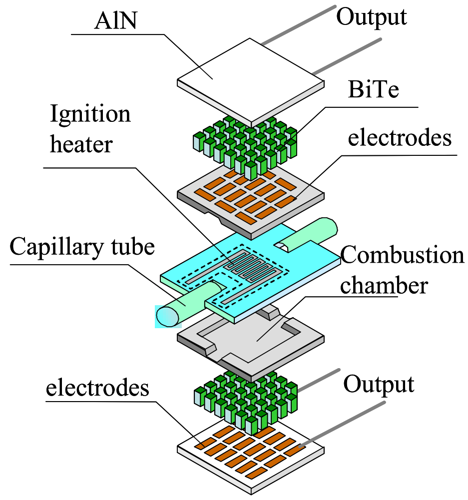

Fig 2: Structure of the TE generator

Table 1 Specification of the TE generator

Size of the TE generator	14 mm × 10 mm × 6.8 mm (0.955cm^3)
Size of the combustor	14 mm × 10 mm × 1.8 mm (0.252cm^3)
Size of the combustion chamber	8 mm × 8 mm × 0.4 mm × 2 (0.05cm^3)
Catalyst and support	Pt-TiO_2
p-type TE element	Bi_2Te_3-Sb_2Te_3
n-type TE element	Bi_2Te_3-Bi_2Se_3
Number of pn couples	34
Size of each TE element	0.95 mm × 0.95 mm × 2mm

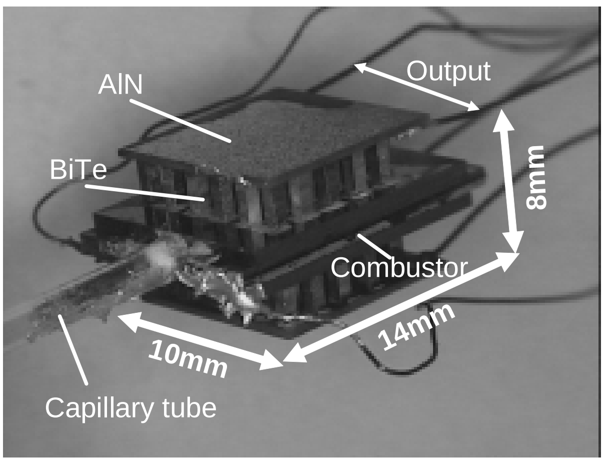

Fig 3: Prototyped TE generator

4. EXPERIMENT

In the experiment as shown in Fig. 4, methanol was used as fuel, because it will become a common fuel for portable fuel cells. A gas mixture of methanol and air was supplied by air-bubbling in a methanol tank. This gas mixture was fed into the combustor and reacted in the catalyst-coated chamber. By using heat generated by the combustion, the TE module produces electric power. The TE module consists of 34 BiTe elements with an area of 8 mm × 8 mm. Two TE modules were attached to both sides of the combustor using silicone grease. The thermocouples were held in place by inserting 1 mm thick Al plates as shown in Fig. 6. The two TE modules were connected electrically in series, and a variable resistor was connected to the TE modules. In order to measure the voltage applied to this load, a voltmeter was connected in parallel. Two heat sinks with a fin length of 20 mm were attached to the TE modules as shown in Fig. 5.

To make the flow rate of methanol constant, the flow rate of air was fixed at 200 sccm. In this condition, the flow rate of methanol was 9.7×10^{-4} mol/min, corresponding to a combustion power of 12.3 W. The flow rate of methanol was calculated measuring the weight change of the methanol. To keep the flow rate of methanol at constant, the temperature of methanol must be controlled at constant. Otherwise, methanol is cooled down by losing latent heat during vaporization, and the flow rate decreases.

Before the power generation test, exhaust gas was analyzed using a gas chromatography system. As a result, combustion efficiency of over 90 % was achieved by this combustor.

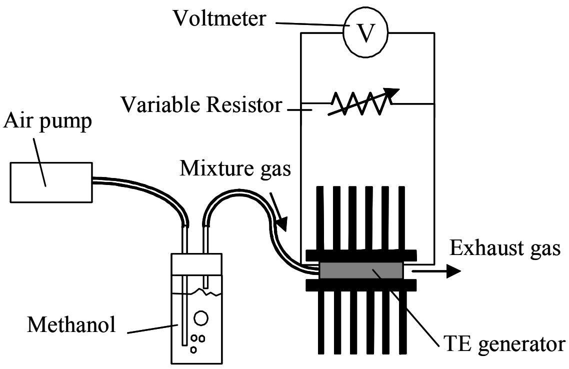

Fig.4 Experimental setup

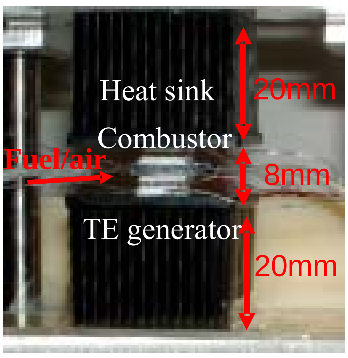

Fig.5 Experimental setup showing the TE generator, combustor and heat sink

In this experiment, the conditions were changed in step-wise as shown in Table 2. At first, the combustor was ignited without connecting a load (resistance) to the TE elements (1). After *Th* reached the maximum temperature, the TE elements were connected to the load. And then, by changing the resistance of the load, the condition where the output was maximized was found (2). Next, *Th* and *Tc* were measured with the TE modules open (3). To confirm the long term stability, a load of 4 Ω was connected, and the output and *Th* were measured (4). Finally, starting-up was confirmed again after stopping combustion (5).

Figure 7 shows the change of *Th* and *Tc* of both TE modules and output voltage. After the air pump was turned on, *Th* increased over 140 ºC, and the load was connected to the TE elements (1). The reason why the top side temperature was higher than the bottom side one is that the top TE module was warmed more than the bottom one by convection. The starting-up time was about 5 sec., which is enough short because the generator is small. The conversion efficiency reached the maximum of 2.39 % at an output power of 294 mW. At that time, the load was 8 Ω , and *Th* and *Tc* were 130 ºC and 50 ºC in average, respectively. The achieved efficiency (2.39 %) is about three quarters of our goal (3.2 %).

When the TE modules were opened (no load was connected), average *Th* rose up to 137 ºC from 125 ºC, since no electric power was consumed by the load (3).This result indicates that the efficiency given by Eq. 2 depends on power consumption, because *Th* varies with output power.

Table 2 Experimental conditions

Step	Air pump	Resistance	Check point
1	on	Infinite	Starting up
2	on	Variable	Maximum power
3	on	Infinite	Heat consumption
4	on	Constant	Stability
5	off→on	Constant	Starting up

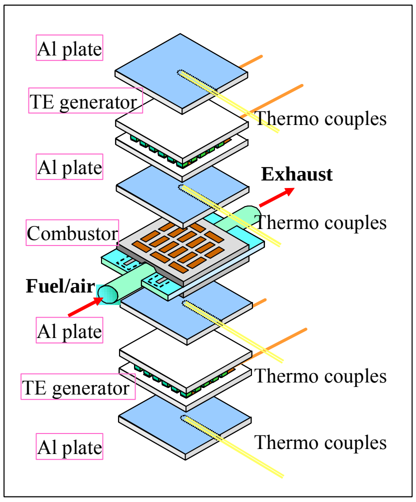

Fig.6 Exploded drawing of the tested TE generator

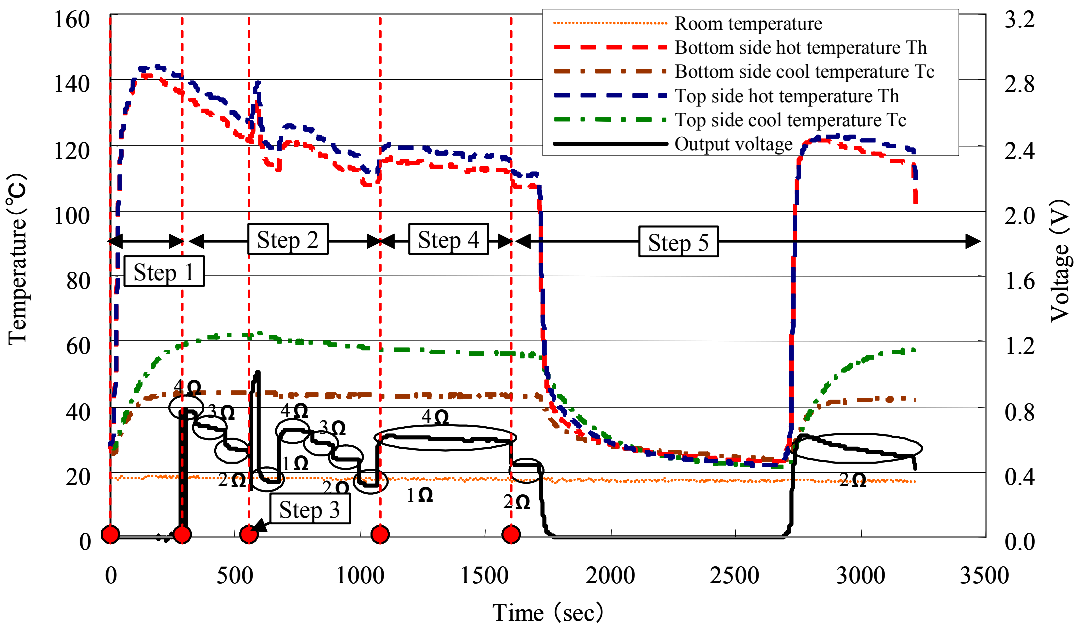

Fig.7 Temperatures (Th and Tc) and output voltage of the TE generator

Next, the stability of output was tried to be confirmed at a constant resistance of 4 Ω (4). The output power remained close to 100 mW, but it decreased gradually. Because the temperature of methanol was not controlled at constant, the concentration of methanol in air decreased due to the cooling of methanol during vaporization, and the stability was not sufficiently confirmed in this experiment. In the next experiment, long-term stability will be checked using a improved setup.

7. CONCLUSION

For applications where exhaust heat is also useful, we developed the components and system of a thermoelectric (TE) generator with a catalytic combustor.

We performed the generation test by attaching two TE modules to the combustor and using methanol as fuel. The conversion efficiency reached 2.39 % at an output power of 294 mW with a 8 Ω load connected. At that time, the average temperature at a hot and cold junction were 130 ºC and 50 ºC respectively. The conversion efficiency approached practical level, but still lower than our goal (3.2 %). The starting-up time was shorter than 5 sec after fuel was supplied. This is enough short because the generator is small. In the next experiment, long-term stability will be checked using a improved setup.

ACKNOWLEDGEMENT

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