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Further Stabilization and Power Density Improvement of Stack-Type Thermoelectric Power Generating Module with Biphasic Medium by Using Various Flexible Metals as Electrodes

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Abstract

In order to realize further stability of a stack-type thermoelectric power generating module (*i.e.* no electrical connections inside), flexible materials of metal springs and/or rods having restoring forces were installed between lower-temperature-sides of thermoelectric elements. These flexible materials were expected to play three important roles of interpolating different thermal expansions of the module components, enlarging heat removal area and penetration of any media through themselves. Then, a low-boiling-point medium (*i.e.* NOVEC manufactured by 3M Japan Ltd.) was also applied for a high-speed direct heat removal via its phase change from the lower-temperature-sides of the thermoelectric elements in the proposing stack-type thermoelectric power generating module. No electrical disconnections inside the module were confirmed for more than 9 years of use, indicating further module stability. The power generating density was improved to about 120 $mW \cdot m^{-2}$ with SUS304 springs having 0.7 mm diameter. Increasing power generating density can be expected in terms of suitable selection of flexible metal with high *Vickers hardness*, cavities control on the spring surface, more vigorous multiphase flow with adding powders to the medium and optimization of the module configurations according to numerical simulations.

Keywords

Thermoelectric Power Generation, Stack-Type Module, Flexible Section, Biphasic Medium, Phase Change, Multi-Phase Flow, Heat Transfer Enhancement, Module Stabilization

1. Introduction

In our previous work, flexible sections were installed to a stack-type thermoelectric power generating module so as to realize reliable electrical connections inside over an extended time period under varying heat source temperatures [1] [2]. Moreover, a biphasic medium composed of an underlying water-insoluble/extremely-low-boiling-point medium (*i.e.* NOVEC manufactured by 3M Japan Ltd.) [3] in small quantity and upper-layered water in large quantity [4] was applied for enabling high-speed direct heat removal via NOVEC phase change from lower-temperature-sides of the thermoelectric elements, expecting a larger-temperature-difference between one side and another of each thermoelectric element (*i.e.* higher outputs) [1] [2]. As the results, no electrical disconnections inside the module could be sustained for more than 7 years of use, confirming the module stability, and the power generating density was improved about two thousands fold, compared to that without flexible sections and with only water instead [1]. However, one defect still remains that compensation of void volumes due to shrinkages of the module constituting materials when heating temperature is decreased since the applied flexible material of copper wools has no restoring force [1] [2].

In this study, two flexible materials of metal springs and/or rods having restoring forces were applied to the stack-type TEG module for highly reliable electrical connections of the module components with different thermal expansion coefficients and high-speed direct heat removal from lower-temperature-sides of thermoelectric elements (*i.e.* higher power generating density).

2. Experimental

2.1. Experimental Apparatus

Figure 1 shows schematics of unit segment from the thermoelectric power generating apparatus (the prototype TEG module) with two ideas of the installation of flexible sections and the utilization of a biphasic medium composed of an underlying water-insoluble/extremely-low-boiling-point medium of NOVEC in small quantity and upper-layered water in large quantity [4]. It should be appreciated that heat removals from the lower-temperature-sides of the thermoelectric elements take place owing to medium boiling heat transfers *i.e.* a large temperature-difference between one side and another of each thermoelectric element, which is well-known directly proportional to a figure of merit in TEG module [5] [6] [7], and the power generating densities must be improved.

The TEG module was mainly composed of three couples of the thermoelectric elements of P-type $Bi_{0.3}Sb_{1.7}Te_3$ and N-type Bi_2Te_3 merchandised by Toshima

Manufacturing Corporation, Ltd., of which the lower-temperature-sides were electrically connected with the flexible sections, and four solid copper rectangular column heaters engulfed by a thin layer of metal wools for assured connection between the higher-temperature-sides of the thermoelectric elements electrically. Each thermoelectric element had a height of 9.0 mm, a width of 4.0 mm and a depth of 9.0 mm. The exterior frame of the module was made of acrylate resin, which has an electrical insulating property, in order to avoid any unforeseen circulatory shunt among the components of the TEG module.

Meantime, the whole experimental apparatus of the prototype TEG module is perfectly the same to our previous work (see **Figure 3** in the literature [1]).

2.2. Experimental Procedures

Firstly, predetermined numbers of metal springs and/or rods as flexible materials were installed into the three flexible sections. Wools were also utilized by way of comparison. **Table 1** summarizes the flexible materials tested.

Secondary, a set amount of NOVEC/water biphasic medium was poured into the upper medium bath. Thirdly, a copper plate cover was placed on the top of the medium bath, making the apparatus airtight. Fourthly, four solid copper rectangular column heaters were submersed in hot water at 353 K, and cooling water at 293 K was circulated through a copper pipe attached to the copper plate cover. Finally, the generated voltage was logged for 60 min by an oscilloscope (NR-500, KEYENCE Corporation, Japan) with an uptake-rate of 1.0 MHz. All experiments were carried out more than three times, and a mean voltage from raw data was converted into power generating density.

Here, favorable properties of NOVEC merchandised by 3M Japan Ltd. [3] with indispensable properties in **Table 2**.

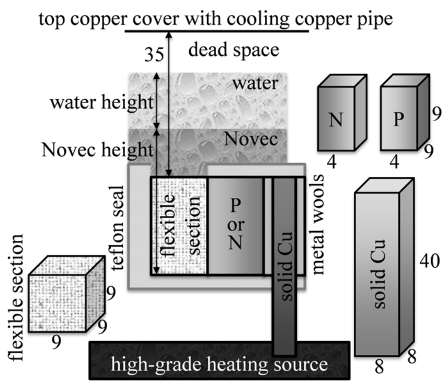

Figure 1. Schematic of unit segment of prototype TEG module.

Table 1. Materials into flexible sections.

materials into flexible sections	shapes
copper	springs
silver	springs
SUS304	wools, rods, springs

Table 2. Properties of NOVEC comparing with water.

	NOVEC		water
boiling point [K]	307	<<	373
density [-]	1.4	>>	1.0
latent heat of vaporization [$kJ \cdot kg^{-1}$]	142	<<	2254
specific heat [$kJ \cdot kg^{-1} \cdot K^{-1}$)]	1.3	<<	4.2
extremely low water solubility (<50 ppm) extremely low corrodibility			

However, NOVEC is too expensive about \$100 kg^{-1}, and it has quite low convective heat transfer coefficient about one sixth to water in spite of pretty unique properties of water-insolubility, anti-corrodibility and a high specific density as shown in Table 1. Then, a biphasic medium of the underlying NOVEC layer and the upper layered water was justifiably emerged so as to reinforce and capitalize on each strength of NOVEC and water. Concretely, more vigorous flow ascribable to condensations of NOVEC vapor bubbles in the upper layered water was expectable, and this might be directly linked to high-speed entropy discharge outside the proposing stack-type TEG module due to convective heat transfer by water having a high convective heat transfer coefficient. Furthermore, NOVEC/water biphasic medium provides reductions of initial cost and weight of the proposing stack-type TEG module since NOVEC has costliness as described-above and specific density of 1.41 above that of water as shown in Table 2.

3. Results and Discussion

3.1. Selection of Best Engulfing Metal Wools to Heater

Figure 2 shows representative time trends of power generating densities from the TEG module with various metal wools thinly engulfed to the four solid copper rectangular column heaters. It can be seen that the power generating densities were decreased towards time elapsed, and approached their respective constant values after individual transition. Additionally, all the experiments indicated the similar tendencies described above, and great repeatability is revealed. Then, their steady values, which are denoted by a transparency gray color region in Figure 2, are shown in the following figures together with their error margins, and discussed in details.

It is also seen that champion power generating density is emerged with using the silver wools, resulting from its highest thermal conductivity and non-corrodibility. Then, the silver wools were used as engulfing metal wools to the heaters in the following experiments.

3.2. Time Trend of Output Voltage with Various Shapes of SUS304 Flexible Material

Figure 3 shows time trends of output voltages with the SUS304 flexible materials in wool, rod and/or spring shapes under changing the heating temperature at 353 K and 293 K with irregular intervals.

It can be seen that the output voltages with wools and rods are slightly decreased towards time elapsed (*i.e.* compensation of void volumes due to shrinkages of the module constituting materials does not work so as being up to expectations). On the contrary, SUS304 springs are excellent without any performance deteriorations. Basically, the restoring forces are concluded one of the most outstanding features for the adopted flexible materials in the proposing TEG module.

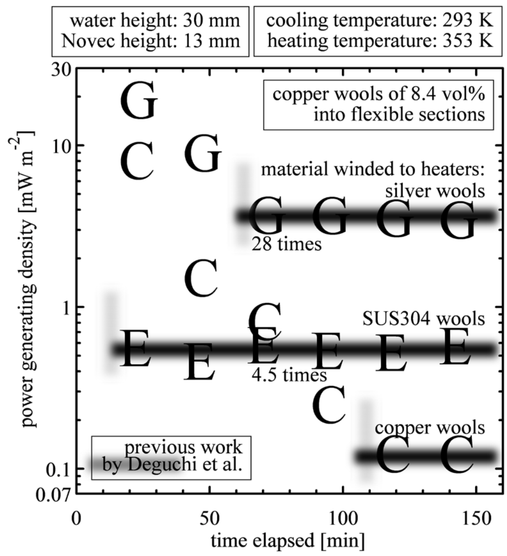

Figure 2. Time trend of power generating density with various metal wools to heaters.

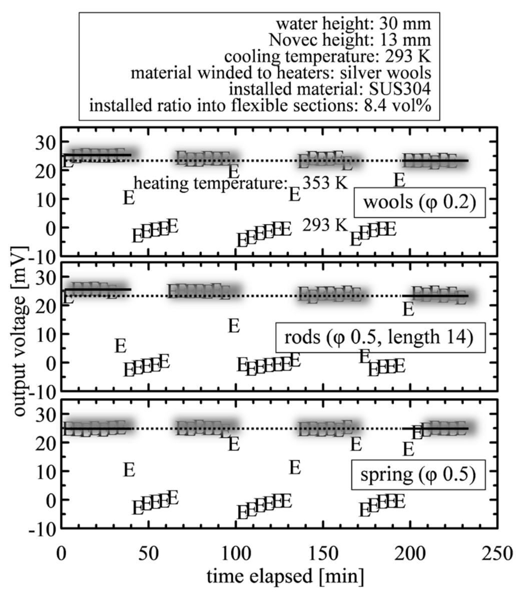

Figure 3. Time trends of output voltages with various shapes of SUS304 flexible material.

3.3. Power Generating Density with Various Shapes of SUS304 Flexible Material

Table 3 shows power generating densities with various flexible materials in spring shape under constant conditions of 30 mm water height, 13 mm NOVEC height, heating temperature 353 K, cooling temperature 293 K and installed spring ratio into flexible sections 8.4 vol%.

It is seen that increasing power generating density can be obtained with increasing metal wire diameter without any exceptions, resulting from a stretching force of the flexible material. Here, values of Vickers hardness are 46, 26, 150 HV for copper, silver and SUS304, respectively. The reason why superior power generating densities reveal with using SUS304 spring might be the highest Vickers hardness among them. Almost the same power generating densities with copper and silver springs, which have quite different Vickers hardness values, are due to quite high and almost the same thermal conductivity at around 400 $W \cdot m^{-1} \cdot K^{-1}$.

Additionally, increment of flexible metal wire diameter leads to decrements of number of springs, heat transfer area and contacting area between flexible material and thermoelectric elements, affecting inferior effects for thermoelectric power generating density. Though increasing the installed spring ratio makes all the values up, medium penetration through the flexible sections must be disturbed, simultaneously, and then the power generating density might be cut down. Briefly, more experiments and numerical simulations [8]-[14] are still needed for optimizing this system since all the parameters interact each other. One idea to keep contacting area between flexible springs and thermoelectric elements constant is to glue "certain fixed installing basements" on the both sides of metal springs, regardless of operating parameters.

3.4. Thermoelectric Power Generation with and without SUS304-Plates Together with SUS304 Springs

Figure 4 shows overall resistance throughout the whole module with and without the "installing basements" of SUS304-plates on the both sides of metal springs.

As expected, the resistance can be decreased due to SUS304-plates, providing areal contacts between flexible springs and thermoelectric elements. **Figure 5** shows power generating densities for these occasions depicted in **Figure 4**.

Table 3. Power generating densities with various flexible materials in spring shape.

material	diameter		
	0.5 mm	0.6 mm	0.7 mm
copper wire	20.6 $mW \cdot m^{-2}$	21.0 $mW \cdot m^{-2}$	21.6 $mW \cdot m^{-2}$
silver wire	22.4 $mW \cdot m^{-2}$	22.7 $mW \cdot m^{-2}$	22.7 $mW \cdot m^{-2}$
SUS304 wire	39.0 $mW \cdot m^{-2}$	40.3 $mW \cdot m^{-2}$	40.9 $mW \cdot m^{-2}$

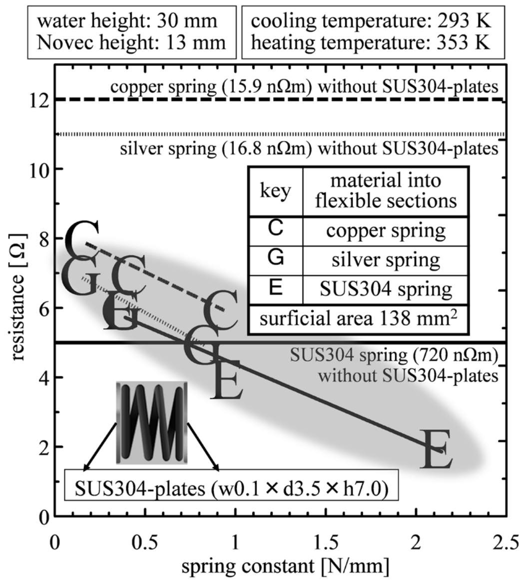

Figure 4. Resistance throughout module with and without SUS304-plates.

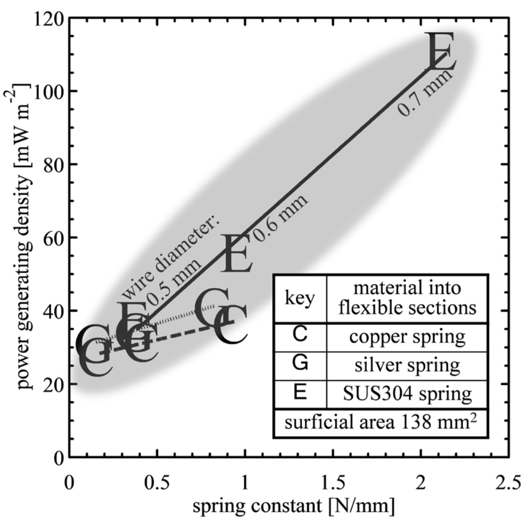

Figure 5. Power generating densities with respect to spring constants.

It is seen that the power generating density is directly proportional to the spring constant, meaning that higher Vickers hardness of the springs for the flexible material is favorable to this system. Though cemented carbides are considered promising flexible materials, cost performance, manufacturing easiness, properties such as chemical stability, thermal conductivity, electric conductivity and affinities towards other constituents must be also important articles for its suitable selection. The campion power generating density was about 120 mW·m^{-2} with SUS304 springs with 0.7 mm diameter. Assuming that the power generating density was extrapolatively proportional to the spring constant, the power generating density can be doubled up to about 250 mW·m^{-2}.

Meanwhile, no electrical disconnections inside the module has been observed after starting this experimental work since the beginning of 2009 fiscal year, resulting to more than 9 years of use under a huge variety of experimental operations including on/off operations (*i.e.* steep temperature changes). Hence, it can be concluded that the installation of the flexible sections must be quite helpful for the TEG modules stabilizations.

4. Conclusions

For further stabilization of a stack-type thermoelectric power generating module with using NOVEC/water biphasic medium and flexible sections, flexible materials of metal springs and/or rods with restoring forces were installed between lower-temperature-sides of thermoelectric elements. The flexible SUS304 springs successfully achieved no electrical disconnections inside the module after starting this experimental work since the beginning of 2009 fiscal year, leading to more than 9 years of use under a huge variety of operations including on/off operations (*i.e.* steep temperature changes).

The power generating density was improved to about 120 $mW \cdot m^{-2}$ with SUS304 springs having 0.7 mm diameter. Assuming that the power generating density was proportional to the spring constant, the power generating density might be upgraded to about 250 $mW \cdot m^{-2}$.

Increasing power generating density can be expected in terms of suitable selection of flexible metal with high *Vickers hardness*, cavities control on the spring surface, more vigorous multiphase flow with adding powders to the medium and optimization of the module configurations according to numerical simulations.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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