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Performance Evaluation of Thermoelectric Materials: A Case Study of Orthorhombic Tin selenide (S_nS_e)

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ABSTRACT: *Designers often face the predicament of non-standardized and poor performing materials for thermoelectric module manufacturing. Other than analytical means, the only method to evaluate the performance of thermoelectric materials would be through experimental means. This work studies the experimental approach employed in performance investigation of thermoelectric materials using Orthorhombic SnSe crystals as a case study. The result obtained reveals the high thermoelectric conversion efficiency of orthorhombic crystals, and that they can operate as both low and high temperature thermoelectric material.*
Keywords: *Conversion Efficiency, Seebeck, Temperature, Thermoelectric generator, Tin selenide.*

I. INTRODUCTION

The conversion of heat energy into electrical energy, occasioned by the difference in temperature is done by a device called thermoelectric generator (TEG). This device is usually in a solid state, and uses a process known as Seebeck effect which may equally be generally referred to as an effect due to temperature gradient. Charge carriers (electrons/holes) are the working flux in a thermoelectric power cycle, which obeys the fundamental laws of thermodynamics. Thermoelectric generators consist of fixed, immovable and highly reliable components that require no maintenance. Their suitability for equipments with low power needs which are located at inaccessible places that are devoid of public power supply (such as vacuum or space, mountain tops, etc.), is emphasized by their high durability rate and long lifespan which sets them apart from other devices. Major challenge of TEG is to find materials with high conversion efficiency in other to boost its output.

II. OBJECTIVES OF THE STUDY

This research, which focuses on the experimental analysis of the conversion efficiencies of Orthorhombic Tin selenide as a thermoelectric material, will be guided by the following objectives:

1. to identify if the thermoelectric materials is reliable in terms of its time of steady-state operation;
2. to identify if it can operate at very high temperatures;
3. to identify if it can operate at both low and high temperatures;
4. to identify the rate at which electrical energy changes with variations in temperature.

III. EXPERIMENTAL OVERVIEW

The junction temperatures and attached load resistance are the two sections that are controlled to change the performance output values when a thermoelectric module is operated as a generating device. The performances are regularly assessed to ensure that the state of these temperatures are constant, indicating that the rate of heat transfer is at a net value and stable with time. The hot side is usually equipped with a heat source that can take various forms. The commonest method is a surface to surface solid heating using plate or flat heaters (Faraji and Akbarzadeh, 2013). Instead of using prefabricated heaters, Cartridge heaters or resistance wire are sometimes used. These are fixed in metallic blocks of high conductivity (mainly aluminum and copper). All the surfaces of the heaters other than the one in contact with the module are properly insulated. In order to achieve steady state conditions, the heat source is powered by a stable source (such as a DC power source) so as to maintain continuous and constant power supply.

There has to be a constant rejection of heat at the cold end of the module to avoid both junctions of the module from eventually reaching thermal equilibrium which would disrupt power generation. The removal of heat on the cold side of the module is achieved by using forced fluid convection cooling. This depends on the amount of heat that needs to be reduced by the module. Forced air convection using fans or air blowers would be sufficient for smaller heat wasting rates while liquid cooling using water through cooling jackets or heat exchangers are used when a larger amount of cooling is needed.

Ethylene glycol mixtures are the commonest liquids used to achieve effective cooling of water temperatures below freezing point. It is imperative to have constant flow rates of the cooling fluids as a precondition to achieving and maintaining steady state conditions. Forced air convection flow rates can be maintained or varied at a constant value by manipulating the input power to the fans. Liquid cooling is mainly realized by a secondary heat exchange method in which the absorbed heat from the intermediary fluid is discharged in the ambient using a heat pumping or refrigeration process. Recirculating chillers or bath temperature controllers are used to achieve such conditions. These instruments are electronically designed and controlled either by using internal or external control systems that usually employ a form of proportional-integral-derivative (PID) control (Anatychukl and Havrylyuk, 2011), aimed at maintaining a desired temperature for the circulating fluid. Variable speed or positive displacement pumps are used to supply flow of liquid. Bypass lines with adjustable valves are used to control the flow rate of fluid when using positive displacement pumps (Huang et al, 2000).

Electronic loads can be used to manipulate the load resistance value attached to the TEG. The basic functions of the electronic loads are to test power supplies, fuel cells and power generating devices. When fixed to a power producing device, the electronic load draws either a constant amount of voltage or current. The device can equally replicate a constant resistance value with precise control over prolonged testing periods.

IV. EXPERIMENTAL SETUP

The setup used in this study was designed to evaluate the performances of Tin selenide module. The test stand accommodates thermoelectric modules with areas of up to 30×30mm^2. Figure 1 shows the setup of the test stand connected to a switchable circuit that consists of an electronic load and a power supply while Figure 2 shows a photograph of the test stand without the side insulation pads. The electronic load was a BK8500 Precision model, capable of testing up to 300W of power from a source. The unit can measure from 0 to 200V of voltage and 0 to 30A of current.

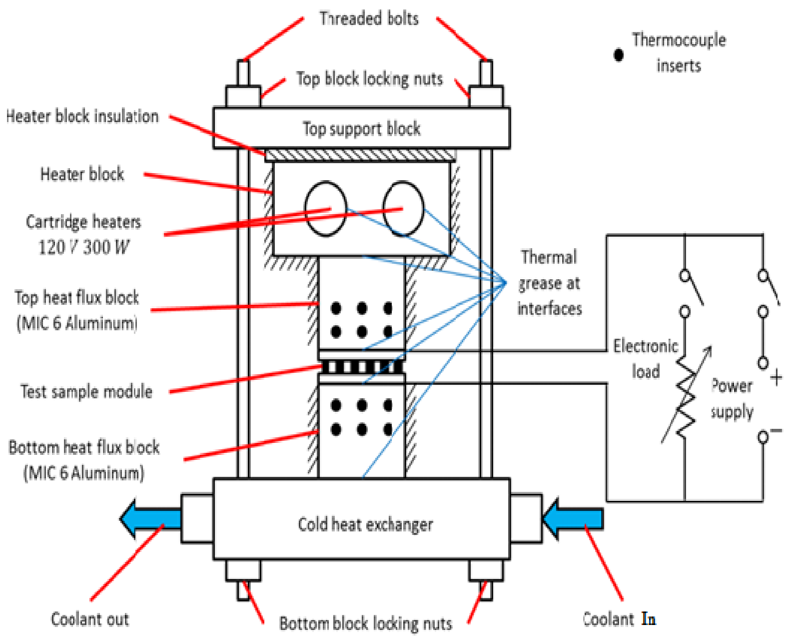

Fig.1: Schematic of Experimental Setup

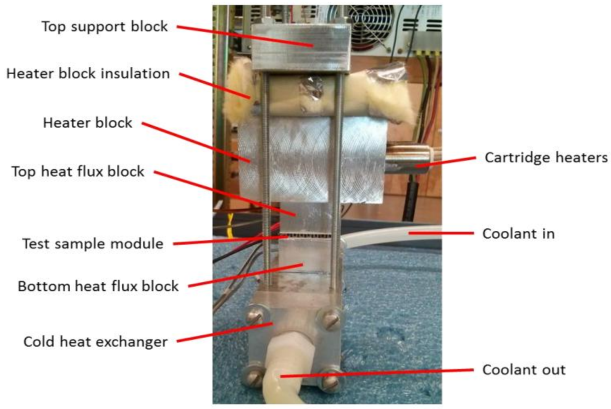

Fig.2: Photograph of Test Stand (without Side Insulation)

A heater block that consists of two in-built cartridge heaters supplied the heat. Omega Engineering Incorporated was the producer of the cylindrical cartridge heaters (Part no: CSS-403300/120V). They had dimensions of 15.8mm in diameter and 88.9mm in length. Each cartridge heater was rated to have up to 0.078W/mm^2 of power density, with a total of 300W at a maximum voltage of 120V. The resistance coils were installed inside a rust proof sheath with a greatest temperature of 667ºC, which was far more than the requirements of the experiments. Stainless steel sheaths were not needed due to the cartridge heaters would not be exposed to any ionized or corrosive fluids. The power supply connected to the cartridge heaters was a TDK-Lambda EMS80-60 model (refer to Appendix A) with an output of up to 80V and 60A of DC power for a maximum of about 5kW of power.

The heat dissipation of the module was accomplished by utilizing a Thermo Scientific NESLAB RTE 7 recirculating chiller. The chiller is made up of a refrigeration system, circulating pump and a microprocessor temperature controller. The unit used had a temperature range capacity of between -25ºC and 150ºC. The capacity of the pump was 15liters/min at 0ºC, and the operating fluid used was 50:50 glycerin/water with freezing and boiling temperatures of -22.8ºC and 106ºC, respectively. An internal 800W heater was used alongside an embedded PID control to maintain the recirculating fluid at a desired working temperature (set point). The heat liberated from the module was absorbed by the working fluid via a one-pass, rectangular channel heat sink.

The heater and cold side heat sink both sandwiched the test sample with respective heat flux blocks in between. These heat flux blocks were machined from MIC 6 aluminum alloy with an approximate thermal conductivity of 142Wm^{-1}K^{-1}. Each block had a contact surface area of 30×30mm^2 and height of 20mm. There were six thermocouple inserts in each block with three slots on one horizontal level and another three on another horizontal level with a perpendicular distance of 5mm between each row (center to center). K-type thermocouples clad in standard stainless steel sheathing were fitted with these inserts. Each insert had a diameter of 2mm and a depth of 20mm. The heat flux blocks were insulated at all surfaces other than those in contact with either the cold or hot sources and the module's surfaces using fiberglass held together by reflective tape. These heat flux blocks functioned in two ways. The first was to measure the heat flux that occurred at the particular junctions of the module and the second was to measure the junction temperature of the modules through a linear method of extrapolation. It was possible to set values by manipulating the voltage output of the power supply connected to the heater and the temperature set point of the chiller. The control loop was able to maintain the junction temperatures within $\pm 0.1°C$ of the desired values. Simple proportional gain (determined through a series of trial and error) was used since the settling time of the system was not crucial.

V. EXPERIMENTAL PROCEDURE

This section reports the stages involved during the obtaining of performance data on a module being tested. Figure 3 below illustrates the processes involved in obtaining the different data points that would be tabulated or graphed to show the performance of a particular module. The first step in the process was to mount the specific test subject between the heat flux blocks (refer to Figure 1). Since the surfaces of the heat flux blocks had micro cracks and surface imperfections, highly conductive thermal paste was applied onto such regions to reduce thermal contact resistances.

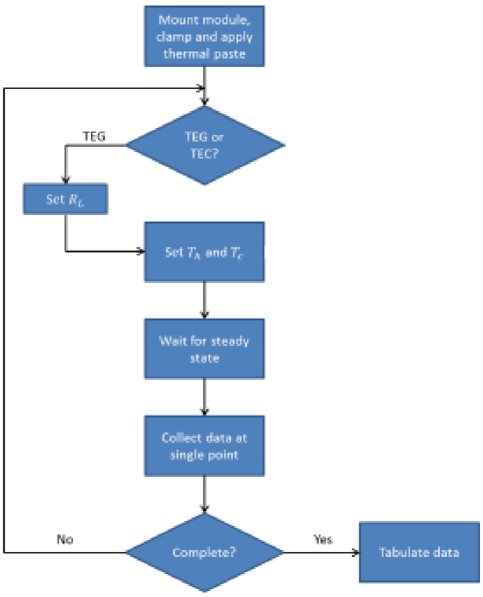

Fig.3: Process Flowchart of Experimental Performance Evaluation

The test stand was then bolted down with the locking nuts using a torque wrench to ensure that all bolts applied equal pressure.

In executing the second step, the selection of TEG was made. In this testing system, the electronic load was fixed to the leads of the module and the load resistance was set to a desired value. In this case, the electronic load acted as a variable resistor. Alternatively, the electronic load was capable of operating in steady state modes of constant voltage, constant current and constant power. The electronic load would draw constant voltage, current or power values respectively from the TEG in these modes.

The additional step was to indicate the junction temperatures to which the heater and chiller would maintain the test sample while it generated power. The control system required between 20 to 40 minutes to achieve steady state conditions.

Larger temperature differences across the module or higher input power values were observed as likely to increase the waiting time. Steady state conditions were approximated by inspecting a waveform chart that shows the temperature readings versus time. When temperature values remained unchanged (within $\pm 0.1°C$) for approximately 2 minutes then steady state conditions were achieved and data collection would start.

The first matched load resistance value was obtained using analytical approach (effective material properties). The load resistance was then varied (increased and decreased) about this initial value until the true matched resistance value (one that yielded the maximum power) was identified. The data acquisition process was repeated until sufficient data points are available for data tabulation.

VI. RESULTS

The geometrical information regarding the thermoelectric elements (i.e. the number of couples and geometric ratio) were physically measured using vernier calipers. The process of obtaining these values was done before assembling the modules. It should be noted that the effective material properties were obtained for one couple on the assumption of similar materials and geometry between each thermoelectric element. TABLE 1 below summarizes the experimentally obtained parameters for Tin Selenide TEG module. The tabular data was reported at five different hot junction temperatures but with the same base cold side temperature. Since the maximum outputs of a module are dependent on temperature, five separate cases of maximum efficiencies were computed based on generated maximum parameter information. The maximum parameters generated are Maximum Power (W_{max}), Maximum Voltage (V_{max}), Maximum Current (I_{max}) and Maximum Efficiency (η_{max}).

Table 1 Effective Parameters for Tin Selenid Module

Criterion	**Symbol (Unit)**	**Set I**	**Set II**	**Set III**	**Set IV**	**Set V**
		T_h= 230°C	T_h= 200°C	T_h= 170°C	T_h=140°C	T_h=110°C
		T_c = 50°C	T_c = 50°C	T_c = 50°C	T_c = 50°C	T_c = 50°C
		∆T=180°C	∆T=150°C	∆T=120°C	∆T=90°C	∆T= 60°C
Maximum Parameters	W_{max} (W)	5.55	4.59	3.62	2.87	2.11
	V_{max} (V)	10.95	9.48	8.00	6.39	4.78
	I_{max} (A)	3.21	3.02	2.82	2.54	2.25
	η_{max} (%)	6.47	6.03	5.58	4.74	3.89

The five different hot junction temperatures used are 230°C, 200°C, 170°C, 140°C and 110°C which corresponded to temperature differences of 180°C, 150°C, 120°C, 90°C and 60°C respectively with uniformed base colds side temperature of 50°C. From fig. 1 below, the output power varied in a linear manner with the changes in temperature difference. At the temperature difference of 180°C, its value was 5.55W. The value decreased progressively as the change in temperature was lowered. Its lowest value recorded was 2.11W at a temperature difference of 60°C. This near linear relationship between the output power and changes in temperature difference is in agreement with the concept of thermoelectric properties being dependent on temperature. When compared side by side with manufacturer's provided performance data for commercial TG12-4-01L Thermoelectric module by Marlow Industries, it is seen that Tin Selenide Module generated better output power at temperature difference 90°C and above; and when compared with HZ-2 Thermoelectric module by Hi-Z, its output power is better across all tested temperature differences. These outstanding values of output power generated will certainly translate to good conversion efficiency.

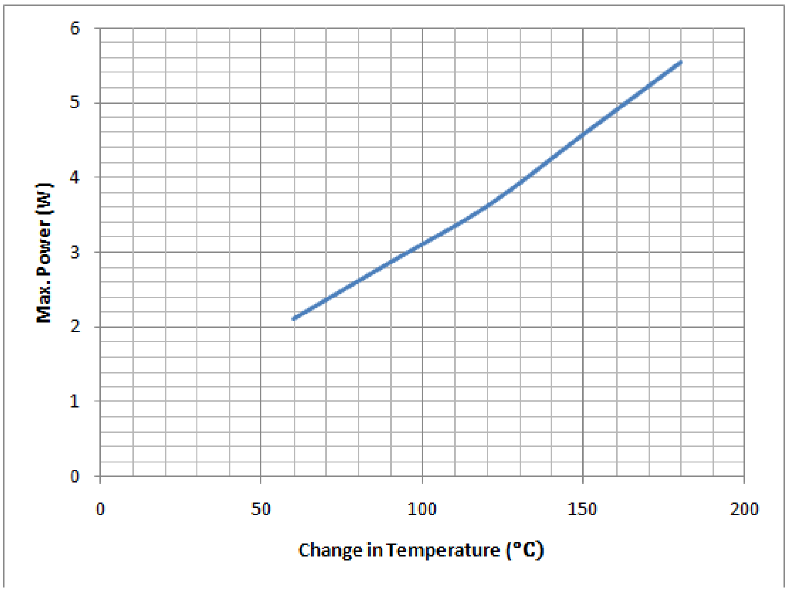

Fig. 4: Max. Power Variation with Change in Temperature for Tin Selenid

Fig.5 below shows the relationship between changes in temperature difference and output voltage. It is seen that this module was able to achieve as high as 10.95Volts at a temperature difference of 180°C, which is a very good fit. As anticipated, the output voltage decreased as the temperature difference decreased but it stood at 4.78Volts at the temperature difference of 60°C, suggesting that the material combination used in this module is suitable for both low temperature TEG and high temperature TEG. This fact is supported analytically by the large Seebeck coefficients, high power factors, and low thermal conductivities of the constituent materials (Sean, 2014). This figure also shows a near linear relationship but is not as steep as fig. 4. When compared with values from commercial TEG modules (TG12-4-01L and HZ-2), it shows a very wide range of difference at same temperature difference all through the tested temperature differences with highest difference of 3.65Volts at temperature difference of 120°C.

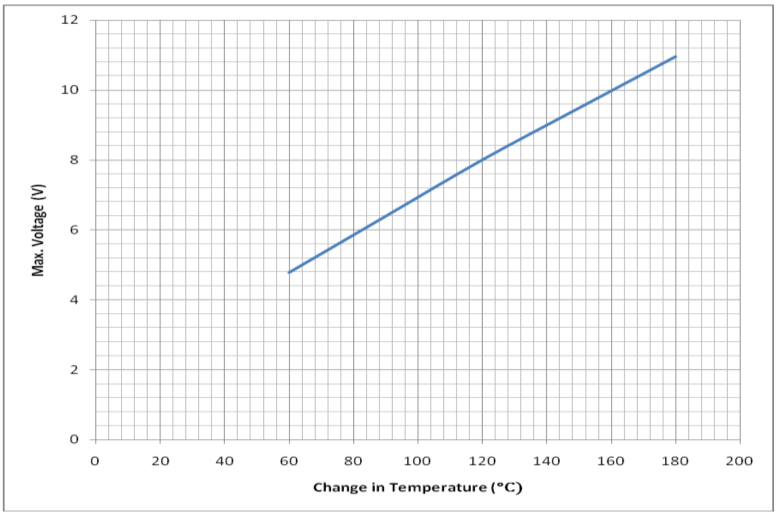

Fig. 5: Max. Voltage Variation with Change in Temperature for Tin Selenid Module

Fig.6 is a plot showing the relationship between temperature changes and output current generated. It is similar in shape to figs. 5 and 4 which agree with ohm's law relationship between current and voltage. Here the maximum value is 3.21A at the temperature difference of 180° C, it decreases linearly as the temperature difference is lowered, and the least measured value was 2.25A at the temperature difference of 60°C. it can be clearly seen that the rate at which the output current decreases with change in temperature difference is very low leading to a useable quantity of current even at low temperature difference. This is a rear characteristic of a thermoelectric material as most of them hardly generate a readable quantity of current at low temperatures (Mahajan, 2013).

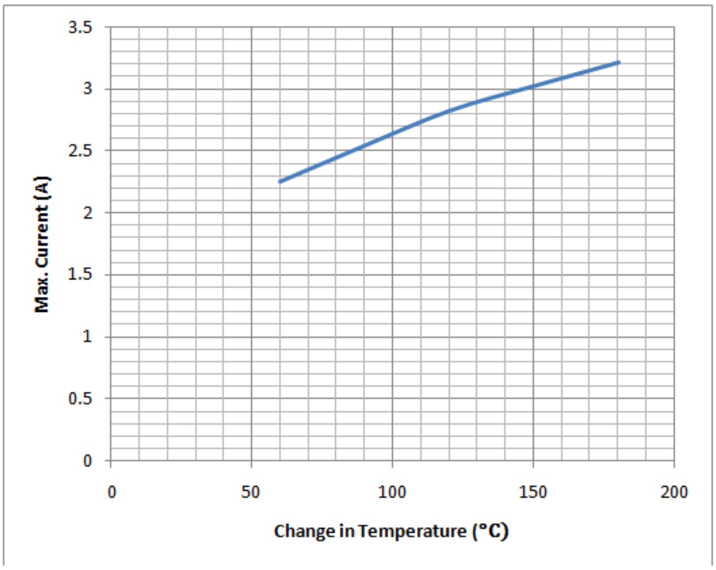

Fig. 6: Max. Current Variation with Change in Temperature for Tin Selenid

The relationship between changes in temperature and efficiency is shown in fig.7. Efficiency is a very important factor in choosing thermoelectric materials, in fact it is the underlining objective of most recent researches in thermoelectric materials including this very one. Low conversion efficiency has been a very big limiting factor for TEGs and has made them almost economically non viable except where there are no alternatives. The efficiency of this module ranged from 6.47% to 3.87% for change in temperature range from 180°C to 60°C. This relatively high conversion efficiency is attributed to high power factor ($S^2\sigma$) along with low thermal conductivity ($\kappa_e + \kappa_l$) of Tin Selenide (Chen et al, 2014). Due to this high power factor the figure of merit for Tin Selenide is about 2.6 at 973K which is highly unprecedented (Guangqian et al, 2015). As expected, the conversion efficiency decreases as the change in temperature is lowered though the rate of decrease is low. Evident of this is the low steep nature of the plot in fig. 7.

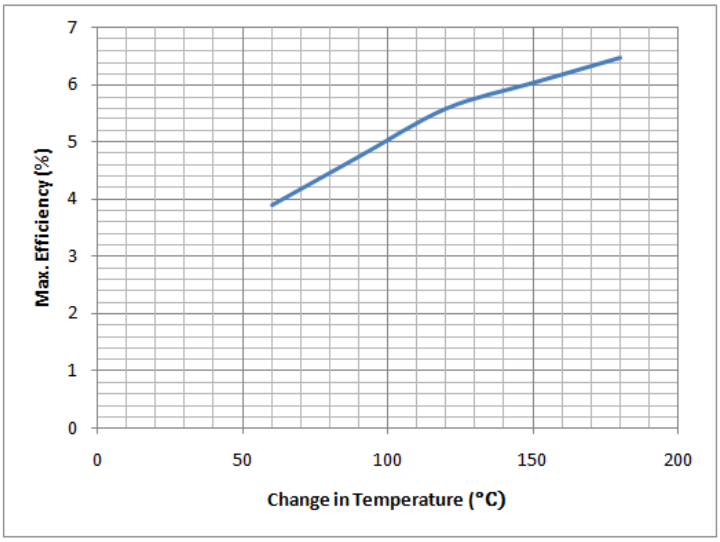

Fig. 7: Max. Efficiency Variation with Change in Temperature for Tin Selenid

VII. DISCUSSION

This research which experimentally investigated the performance of orthorhombic Tin Selenide thermoelectric module had its primary objective of assessing the instantaneous thermoelectric conversion efficiency.

This module displayed unanticipated high conversion efficiency in line with the predictions of (Guangqian et al, 2015) that orthorhombic IV-VI compounds are likely to produce high thermoelectric conversion efficiency due to their unusual high power factor. The maximum conversion efficiency of this module was 4.97% at a temperature gradient of 180°C, generating an electric current of 1.71A and electric voltage of 9.45V. This performance outshines those commonly obtained from regular commercial TEGs at the same temperature reference point. This module was also able to retain its high conversion efficiency at lower temperature gradients. Its conversion efficiency stood at 2.39% at temperature gradient of 60°C. At this point, it generated 0.75A of current and 3.28V of voltage. This shows that this module can be put into use at both high and low temperature reference points.

VIII. SUMMARY

One of the main objectives of this study was to provide designers aiming at implement thermoelectric modules into their designs, alternative materials with high conversion efficiency. The motivation behind this objective was the very poor and non competitive conversion efficiencies of commercial TEGs, which has made them almost irrelevant. A thermoelectric modules made from orthorhombic material have been investigated and it has been found that it have a tremendous conversion efficiencies when compared to available commercial TEGs primarily due to the arrangement of its atoms and its electron mobility. It was discovered that Tin Selenide module has a conversion efficiency which cuts across both low and high temperature gradient reference points. This makes the module's constituent materials very good alternative for use in TEGs especially when the TEG is to be operated across a wide range of temperature difference.

IX. CONCLUSION

It is shown from this study that orthorhombic solids have good thermoelectric conversion efficiency. This good conversion efficiency is attributed to their atomic arrangement and electron mobility which directly influence their power factor. The Tin Selenide module can function both at low and high temperature reference points with output variations relatively stable.

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