

Fabrication of thermoelectric generator using local minerals at Loei Province, Thailand

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Abstract

Local Thermoelectric mineral specimens at Loei Province, in the northeastern part of Thailand, were prepared as powders and bulk solids form by crushing, calcination and annealing, pressure and sintering, cutting and polishing. Mineral samples were used to analyze the composition and phase, determine the thermoelectric property and efficiency, design and construct a thermoelectric generator. Chemical composition and phase identification of powder samples were analyzed by the x-ray fluorescence (XRF) and x-ray diffraction (XRD), respectively. XRF and XRD results indicated that the mineral samples comprised the $\text{Fe}_2\text{O}_3\text{-SO}_3\text{-SiO}_2\text{-others}$, and $\text{Fe}_2\text{O}_3\text{-SiO}_2\text{-CuO-others}$. From the thermoelectric property and efficiency determinations, the p- $\text{Fe}_2\text{O}_3\text{-SO}_3\text{-SiO}_2\text{-others}$ and n- $\text{Fe}_2\text{O}_3\text{-SiO}_2\text{-CuO-others}$ bulks were found to exhibit the thermoelectric figure of merit in orders of 10^{-11} and 10^{-13} K^{-1} , respectively. A fabricated thermoelectric generator made from twenty pairs of p- $\text{Fe}_2\text{O}_3\text{-SO}_3\text{-SiO}_2\text{-others}$ and n- $\text{Fe}_2\text{O}_3\text{-SiO}_2\text{-CuO-others}$ legs that can be provided the open circuit voltage and short circuit current up to 71.7 mV and 0.08 μA for a temperature difference of 38 K at room temperature, respectively.

Keywords: Local minerals, p- $\text{Fe}_2\text{O}_3\text{-SO}_3\text{-SiO}_2\text{-others}$, n- $\text{Fe}_2\text{O}_3\text{-SiO}_2\text{-CuO-others}$, Thermoelectric material, Thermoelectric property and efficiency, Thermoelectric generator

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1. Introduction

Thermoelectric materials depend on properties such as the Seebeck coefficient (S) [1-3], electrical resistivity (ρ) [4-5] and thermal conductivity (κ) [6] for viable use. Efficiency can be calculated from the physical parameters S , ρ and κ , which are employed and given by the thermoelectric figure of merit $Z = S^2/\rho\kappa$, where S^2/ρ is referred to as the electrical power factor (P). The development of materials include high S , low ρ and κ , which lead to the large Z . Thermoelectric performance of materials is usually characterized in terms of their dimensionless figure of merit ZT , where T is the absolute temperature, where large ZT values will be lead to high efficiency. However, only those materials, which possess a $ZT > 0.5$, are usually regarded as thermoelectric

materials [3]. For the Z values of metals, semiconductors, and insulators were about $3 \times 10^{-6} \text{ K}^{-1}$, $2 \times 10^{-3} \text{ K}^{-1}$, and $5 \times 10^{-17} \text{ K}^{-1}$ at 300 K, respectively [7].

The materials for thermoelectric energy conversion are two recognized types of charge carriers, such as positive (p-type) and negative (n-type) charges moving parallel to the heat flow, which can be provided by hot probe experiments. The applications of thermoelectric devices were made from the thermoelectric modules, which consist of two or more materials of n-type and p-type, and connected electrically in series but thermally in parallel [8-9]. The devices are also being used as the thermoelectric generators, coolers, and other applications [10-11]. Therefore, the search for new metal oxide materials with high thermoelectric performance is of wide interest. In this work, thermoelectric minerals have been found at Loei Province, in the northeastern part of Thailand [12-17]. Mineral specimens were prepared in the powders and bulk solids form by crushing, calcination and annealing, pressure and sintering, cutting and polishing. The analysis of chemical composition and phase were investigated. Thermoelectric property and efficiency determinations are presented and discussed. Fabrication and test of a thermoelectric generator made from the local minerals are reported on.

2. Materials and Methods

The experimental procedures including preparation and characterization of mineral samples, thermoelectric property and efficiency determinations, fabrication and test of a thermoelectric generator, and are described as follows.

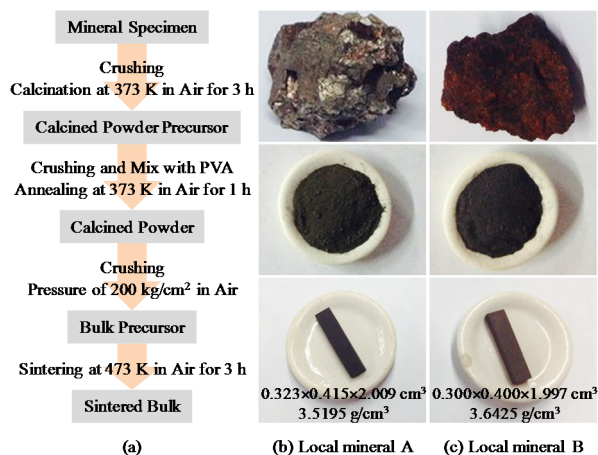


Fig.1 Preparation of powder and bulk samples

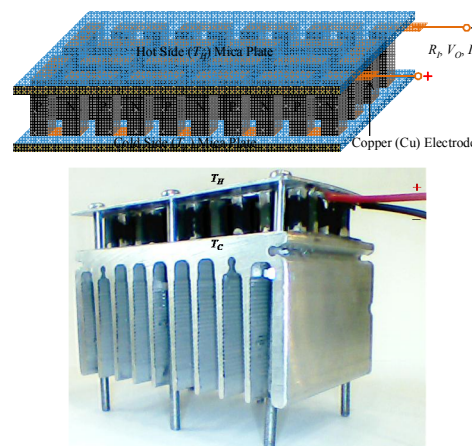


Fig. 2 Design and construction of a thermoelectric generator

Preparation and characterization of mineral samples

Fabrication flow chart for the preparation of mineral samples is shown in Fig. 1(a). Mineral specimens were crushed and calcined at 373 K in air for 3 h. Calcined powders were crushed and mixed with polyvinyl alcohol (PVA) in 5 g: 1 mL ratio and annealed at 373 K in air for 1 h. Annealed powders were crushed and pressed

into bulk solids at the pressure of 200 kg/cm² in air before subjected to sintering stage. Bulk precursors were sintered at 573 K in air for 3 h. Subsequently, sintered bulks were cut and polished for the determination of thermoelectric properties and efficiencies. Mineral specimens, powder and bulk samples are shown in Fig. 1(b) and Fig. 1(c), such as Local mineral A (at Ban Pha Baen, Buhom Subdistrict, Chiang Khan District) and Local mineral B (at Ban Huai Muang, Moo 13, Nadindum Subdistrict, Mueang District). Chemical composition and phase identification of powder samples were analyzed using the X-Ray Fluorescence Spectrometry (Philips PW-2404) and X-Ray Diffractometer (Shimadzu XRD-6100), respectively.

Thermoelectric property and efficiency determinations

Firstly, the thermoelectric sensitivity can be considered by the type of charge carriers and Seebeck coefficient (S), which were determined by the hot probe method. Secondly, electrical resistivity (ρ) was measured by the current and voltage characteristics. Thirdly, the steady state technique was used to find the thermal conductivity (κ). Finally, the thermoelectric efficiency was examined from the material's power factor ($P = S^2/\rho$) and the thermoelectric figure of merit ($Z = S^2/\rho\kappa$).

Fabrication and test of a thermoelectric generator

Fabrication of a thermoelectric generator composed of twenty p-n couples, which were designed and constructed using copper (Cu) electrodes and mica substrates. Dimension of each p-n legs were 0.30 cm width, 0.40 cm length and 1.00 cm height. Two mica plates possessing dimension of 0.10 cm thickness, 4.00 cm width and 8.00 cm length were used as substrates for the thermal translation of the hot and cold sides. Cu sheets of 0.05 cm thickness, 0.40 cm width and 0.90 cm length were attached onto the mica plate using epoxy adhesive to maintain achieve electrical conduction. The p-n legs and Cu sheets on substrates were adhered using silver paste. A designed and constructed thermoelectric generator is shown in Fig.2. The internal resistance (R_i) of the generator was measured. The preliminary test build device was used for thermoelectric power generation. A ceramic resistor was used to apply heat by applying currents to a resistor placed on a hot side (T_H), while a cold side (T_C) was placed on a heat sink to release heat. The other sides of a generator were surrounded by air at room temperature (T_R). The open circuit voltage (V_O) and closed or short circuit current (I_S) of a generator were measured as a function of the temperature difference ($\Delta T = T_H - T_C$).

3. Results and Discussion

The experimental results and discussion presented and reported in this work are chemical composition and phase identification, thermoelectric properties and efficiencies, and test results on a thermoelectric generator.

Chemical composition and phase identification

X-ray fluorescence (XRF) was used to analyze the chemical composition. The concentrations of compounds are given in Table 1. The crushed mineral A included Fe₂O₃ (71.03%), SO₃ (22.38%), SiO₂ (4.21%), and others. The crushed mineral B was composed of Fe₂O₃ (84.95%), SiO₂ (5.88%), CuO (3.78%), and others. Hence, it can be expected that the mineral samples will contain these species composition. From this point onward, the mineral samples A and B will be referred to as Fe₂O₃-SO₃-SiO₂-others and Fe₂O₃-SiO₂-CuO-others containing, respectively.

Table 1 Compounds and concentrations (%) of local minerals

Local mineral A	%	Local mineral B	%
Sodium oxide, Na ₂ O	0.04	Sodium oxide, Na ₂ O	0.17
Magnesium oxide, MgO	0.25	Magnesium oxide, MgO	2.19
Aluminum oxide, Al ₂ O ₃	0.72	Aluminum oxide, Al ₂ O ₃	2.45
Silica, SiO ₂	4.21	Silica, SiO ₂	5.88
Sulfur trioxide, SO ₃	22.38	Phosphorus pentoxide, P ₂ O ₅	0.03
Potassium oxide, K ₂ O	0.11	Sulfur trioxide, SO ₃	0.10
Calcium oxide, CaO	0.42	Potassium oxide, K ₂ O	0.02
Titanium oxide, TiO ₂	0.04	Calcium oxide, CaO	0.20
Manganese oxide, MnO	0.13	Titanium oxide, TiO ₂	0.02
Feric oxide, Fe ₂ O ₃	71.03	Manganese oxide, MnO	0.16
Cobalt Oxide, Co ₂ O ₃	0.23	Feric oxide, Fe ₂ O ₃	84.95
Copper oxide, CuO	0.03	Copper oxide, CuO	3.78
Arsenic trioxide, As ₂ O ₃	0.32	Arsenic trioxide, As ₂ O ₃	0.03
		Molybdenum dioxide, MoO ₂	0.02

Phase identifications were analyzed using the x-ray diffraction (XRD) at room temperature using CuK α radiation, $\lambda=0.15406$ nm. The powder samples were measured in the 2-theta angle range of $20^{\circ} \leq 2\theta \leq 80^{\circ}$ with scanning rate of 0.02°/sec and obtained from the International Centre for Diffraction Data (PCPDFWIN Copyright © JCPDS-ICDD 2003). XRD patterns are shown in Fig.3. The results show the traces of Fe₂O₃, SO₃, and SiO₂ phases in the powder sample A, while the powder sample B was composed of the Fe₂O₃, SiO₂, and CuO phases. XRF and XRD results indicated that the mineral samples comprised the Fe₂O₃-SO₃-SiO₂-others and Fe₂O₃-SiO₂-CuO-others compounds for the local minerals A and B, respectively.

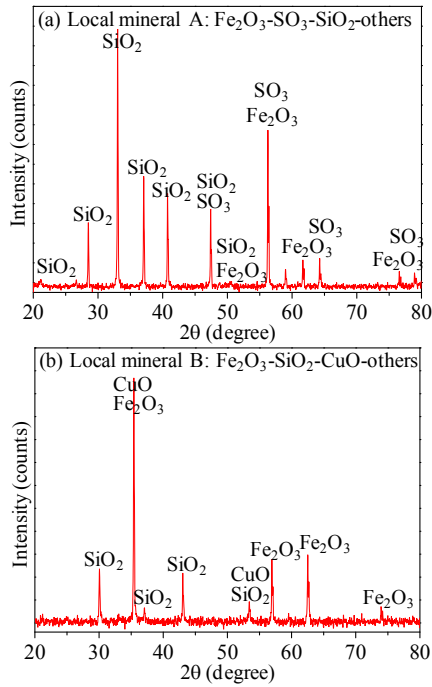


Fig. 3 XRD patterns of local minerals

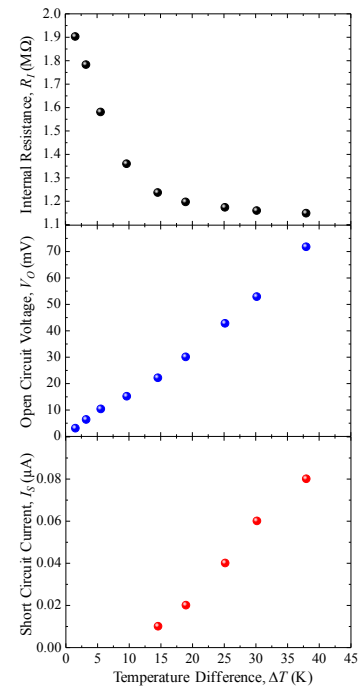


Fig. 4 A fabricated thermoelectric generator and test results

Thermoelectric properties and efficiencies

From the assessment of thermoelectric properties and efficiency, it was found that the $\text{Fe}_2\text{O}_3\text{-SO}_3\text{-SiO}_2\text{-others}$ compound exhibited p-type conduction with $S = 47.42 \pm 1.43 \text{ } \mu\text{V/K}$, $\rho = 0.08 \pm 0.01 \text{ k}\Omega\cdot\text{cm}$, $\kappa = 215.30 \pm 2.82 \text{ W/m}\cdot\text{K}$, $P = 2.70 \pm 0.23 \times 10^{-9} \text{ } \mu\text{W/m}\cdot\text{K}^2$, and $Z = 1.26 \pm 0.12 \times 10^{-11} \text{ K}^{-1}$. The $\text{Fe}_2\text{O}_3\text{-SiO}_2\text{-CuO-others}$ compound was n-type with $S = -76.28 \pm 0.94 \text{ } \mu\text{V/K}$, $\rho = 9.09 \pm 0.80 \text{ } \Omega\cdot\text{cm}$, $\kappa = 336.25 \pm 17.74 \text{ W/m}\cdot\text{K}$, $P = 6.40 \pm 0.41 \times 10^{-11} \text{ } \mu\text{W/m}\cdot\text{K}^2$, and $Z = 1.90 \pm 0.22 \times 10^{-13} \text{ K}^{-1}$. The result of measurement and calculation are summarized in Table 2.

Table 2 The S , ρ , κ , P and Z of mineral samples in air at room temperature

Mineral samples	S ($\mu\text{V/K}$)	ρ ($\text{k}\Omega\cdot\text{cm}$)	κ ($\text{W/m}\cdot\text{K}$)	P ($\text{W/m}\cdot\text{K}^2$)	Z (K^{-1})
p- $\text{Fe}_2\text{O}_3\text{-SO}_3\text{-SiO}_2\text{-others}$	47.42 ± 1.43	0.08 ± 0.01	215.30 ± 2.82	$2.70 \pm 0.23 \times 10^{-9}$	$1.26 \pm 0.12 \times 10^{-11}$
n- $\text{Fe}_2\text{O}_3\text{-SiO}_2\text{-CuO-others}$	-76.28 ± 0.94	9.09 ± 0.80	336.25 ± 17.74	$6.40 \pm 0.41 \times 10^{-11}$	$1.90 \pm 0.22 \times 10^{-13}$

Test results on a thermoelectric generator

For the preliminary test, a fabricated thermoelectric generator was used to generate thermoelectricity in air at room temperature (T_R). The internal resistance (R_i), open circuit voltage (V_o), and short circuit current (I_s) were measured as a function of the temperature difference ($\Delta T = T_H - T_C$). Test results on the generator using local minerals are shown in Fig.4. It was found that the values of open circuit voltage and short circuit current

increased with increasing temperature difference up to $V_O = 71.7$ mV and $I_S = 0.08$ μA for $\Delta T = 38$ K ($T_H = 363$ K and $T_C = 325$ K). While the internal resistance decreased and reached a value of $R_i = 1.15$ M Ω .

4. Conclusion

A thermoelectric generator was designed and constructed using local minerals p-Fe₂O₃-SO₃-SiO₂-others and n-Fe₂O₃-SiO₂-CuO sourced at Loei Province, Thailand. A generator was built with twenty pairs of p-n legs, copper electrodes and mica substrates. This device was used to generate thermoelectricity, and provided an open circuit voltage and short circuit current up to 71.7 mV and 0.08 μA for a temperature difference of 38 K at room temperature, respectively. However, the dimension and number of p-n legs should be further investigated.

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