THERMOELECTRIC PROPERTIES OF LEAD TELLURIDE FILLED IN SILICONE

ABSTRACT

In recent years, much research has been done on thermoelectric materials because of increasing interest in recovering waste thermal energy. Lead Telluride doped with Silicone matrix in liquid form was processed into molecularly aligned fibers under high-voltage electric field. Electrospinning was implemented in this experiment to make molecularly aligned fibers. The length of the manufactured rod was about 50 mm in length and 1 mm in diameter. Electrical resistance and Seebeck coefficient of Lead Telluride doped with Silicon matrix were tested. The electrical property of the thermoelectric Lead Telluride mixture follows ohm's laws. Its resistance is intrinsically related to resistivity, cross sectional area and length. The material also exhibits high Seebeck coefficient. Materials with such properties can be applied in alternative energy applications, HVAC and electronics.

INTRODUCTION

About two-thirds of energy input of electricity generation in the U.S is lost as useless thermal energy during the conversion process [1]. Thermoelectric materials can offer promising breakthroughs due to their ability to recover thermal energy and convert it into electricity. The scientific discoveries of thermoelectric were mainly found during two periods of history. From 1821 to 1851, fundamentals of thermoelectric properties were understood. Intensive experiments were done to understand thermoelectric materials microscopically since 1930s [2]. The properties of thermoelectric materials depend on the Seebeck, Peltier and Thomson coefficients α, Π, and τ. Thomas Johann Seebeck discovered the Seebeck effect in 1823. In a circuit with two dissimilar conductors, a and b, each has different temperatures at junction W and X. If those two conductors are p-type and n-type materials, voltage difference can be induced in the circuit. This effect is called Seebeck coefficient [3]. The equation for Seebeck coefficient is α_{ab}=dV/dT under open circuit [2]. If the temperature at junction W is higher than junction X, α coefficient is positive because a thermocouple ab would produce a clockwise current. The electric potential that is induced depends on the material and the temperature gradient. Hence the equation to calculate the induced electromotive force is $E_{AB}= S_{AB}\Delta T$. S_{AB} is defined as the relative Seebeck coefficient between homogenous conductors, A and B. If more than one pair of conductors are present, then the equation becomes $E_{AC}= E_{AB} + E_{BC} = (S_{AB}+S_{BC})\ \Delta T$. S_{AB} is calculated by subtracting S_B from S_A [3].

The efficiency of thermoelectric materials is determined by the figure of merit. A figure of merit is a quantity that describes performance of the device. A large dimensionless figure of merit value indicates a good thermoelectric property. The efficiency equation is $S = (S^2 *\sigma*T) / k$. s is the Seebeck coefficient. σ is the electrical conductivity. k is the total thermal conductivity [4]. The equation above shows that the square of Seebeck coefficient should increase proportionally with the figure of merit. To ensure that the thermoelectric figure of merit is maximized, a large thermopower which is an absolute value of the Seebeck coefficient S, high electrical conductivity σ, and low thermal conductivity k are required [5]. Furthermore, the thermal conductivity is increased by adding volume fraction of fillers to form an interconnected conductive network [6]. To confirm that the Seebeck coefficient is large, only a single type carrier should be present. Both charged carriers would move to the cold end and cancel out the induced Seebeck voltage if mixed n-type or p-type conduction is present [5]. The thermoelectric power factor PF = $S_2\sigma$ ($Wm^{-1}K^{-2}$) is another wayof determining thermoelectric property other than Seebeck coefficient. The thermoelectric power factor is used for application on energy generation because it indicates the quantities of electric power [7].

Lead Telluride is a semiconductor that has one of the best thermoelectric properties [9]. It has a high melting point, good chemical stability [8] [11]. PbTe is one of the best solid-state thermoelectric materials, and its temperature range is 323-900 K in energy generators [9]. It has a narrow band gap of 0.31 eV at 300K, face centered cubic structure (FCC), and large exciton Bohr radius (46 nm) which makes an ideal material to observe its properties and behaviors under quantum confinement conditions [9] [10]. By utilizing this material, thermal energy can be converted into electric energy. Lead Telluride is used for thermoelectric power thermo-generation for temperature range 500-900 Kelvin [11]. The Seebeck effect is tested in this experiment and it relates the electrical potential difference with temperature changes. The thermoelectric property of Lead Telluride has a wide arrange of uses. It can be used to reuse waste heat from refrigeration, power generation and electronics cooling.

Silicone matrix is produced by mixing Lead Telluride with Silicone in liquid form. Silicone is selected for its low thermal conductivity, low chemical reactivity and non-toxicity. Silicones are also very good water repellent. Due to its flexibility and impermeability, Lead Telluride matrix in liquid form can be applied on industrial heat pipes to recover waste thermal energy. Silicone’s hydrophobicity is caused by low weight molecules in the bulk of the material that skim up to the surface because of difference in diffusion density [12]. Under normal steam pressure, silicone rubber shows no signs of deterioration. In solid form, Silicone rubber is able to retain water repellency, dielectric properties and thermal degradation resistance [12]. –[Si-O]x- chain segments provide flexibility and Si-O bonds have high strength. Silicones have low heat release rate, and this property provides Lead Telluride to absorb heat more efficiently.

Lead Telluride mixed with Silicone matrix was manufactured into nanorods using electrospinning. As the liquid was being extruded out of the syringe, the electric field continuously bends and stretches the liquid into very thin shape. An electrical field influences the liquid droplet until it reaches a critical voltage Φ_0 [13]. As results of electrospinning nanometer rods or wires have high surface to volume ratio and enhanced strength [13]. Electrospinning is the ideal method to produce continuous nanofibers due to its versatility. Electrospinning is easy to set up, and fiber diameter is controllable. This process is used widely in various fields. For the development of electronics, catalytic and hydrogen storage systems, nanofibers are manufactured by such process [14]. Nanofibers generated by Electrospinning sizes range from less than 3 nm to over 1μm [15]. The process of electrospinning was first dated back to the 17th century. William Gilbert first discovered Electrospinning in 1600 [16]. In 1745, Bose created aerosol by using high electric potential to a liquid at the end of a glass capillary tube[17]. Lord Rayleigh predicted minimum charge a liquid could carry to overcome its surface tension. If the charge is unstably large, fission takes places and stabilizing effect of surface tension is minimized [18]. John Francis filed the first electrospinning patent in 1900 [19].

MATERIALS AND EXPERIMENTAL METHODS

0.8 grams of Silicone was weighed prior to mixing. Powder form Lead Telluride (0.2 g) was mixed with the liquid form Silicone and transferred to the syringe. The volume of the syringe was 10 mL. As illustrated in Figure 1, a syringe pump was instructed to push the liquid at a constant speed of 0.05 mL/min. A DC power supply was adjusted to deliver 10KV of voltage to the syringe tip. The voltage applied in the electrospinning process is usually ranged from 10 kV to 30 kV [20]. The metal plate was used as a grounded collector for Lead Telluride. Due to electrostatic attraction, a continuous uniform small rod was extruded towards the grounded collector as the syringe pump pushing liquid from the syringe. In this experiment, the grounded collector was a flat rectangular metal plate. The grounded collector is also known as the counter electrode. In addition, other types of possible counter electrode are rotating cylinders, disks, two parallel bars, etc [20]. The Seebeck coefficient of lead telluride doped with silicone was measured with an infrared thermometer and a CHI 600E Electrochemical Analyzer. When measuring the electrical resistance of the sample, scan rate was set at 0.01 volt/second. Initial voltage was set at 0 V, and final voltage was 0.1 V. For measuring Seebeck coefficient, the electrical potential was adjusted between 0-1 V and the runtime was set as 50 seconds.

RESULTS AND DISCUSSION

From Figure 6-9, the temperature depended Seebeck coefficient of lead telluride shows negative Seebeck coefficient which means electrons are the dominant charge carriers [21]. If electrons are diffused from the hot side to the cold side, then the material is n-type. In contrast, electrons tend to diffuse from the cold side to the hot for p-type materials [22]. A large Seebeck coefficient requires the material to be only a single type of carrier. Mixing n-type and p-type will cause both charge carriers to be moved to the cold end and cancel out the induced voltage [23]. Seebeck coefficient is calculated by the ratio between the voltage different induced by the material and temperature gradient. Furthermore, $S = -\Delta V/\Delta T$. Since Seebeck coefficient is temperature dependent, several runs of measurements were performed. Figure 6-9 illustrated the Seebeck coefficient of the thermoelectric material was measured at 319K, 321K, 325K and 331K. The material in this experiment exhibits high absolute Seebeck coefficient values. The Seekbeck effect is directly related with the concentration of lead telluride in the silicone rubber matrix. However, the flexibility of silicone will decrease as a compensation for increasing lead telluride. Proper Proper concentration of lead telluride was considered prior to the experiment.

Figure 2 and 3 showed the relationship between voltage and current. A CHI 600E Electrochemical Analyzer was used to supply an increment voltage of 0.001 from 0 to 0.1 V When current is zero, the voltage is relatively high. Hence, the material behaves like an open circuit. As current increases, the electrical potential decreases as shown in Figure 3. In this case, the material behaves like a close circuit.

Since silicone matrix has the potential to be used as an insulator for the thermoelectric material. Adequate measurements of electrical properties of such material are indispensable. The other reason for testing electrical resistance was that electrical resistivity was one of the factors that determine the figure of merit. The equation of electrical resistivity is $R = \rho*L/A$. The length of the manufactured rod using electrospinning was measured as 50 mm and the diameter was measured as 1 mm. Due to the fact that the highest electrical resistance of the thermoelectric Lead Telluride measured was $5.93*10^{10}$ ohms, the electrical resistivity was calculated to be $9.31*10^{5}$ ohm*m. One of the reasons for selecting silicone rubber is because of its hydrophobicity. Hydrophobicity is related directly with current leakage [24]. Thus, silicone rubber functions as an insulator for the seebeck coefficient experiment. In addition, insulators generally have high seebeck coefficients.

Lead telluride doped with silicone exhibits enhanced Seebeck coefficient and significant increase in resistance. This is due to Silicone's low thermal conductivity and low chemical reactivity. Electrons cannot pass through the lattice structure as easier as other types of materials.

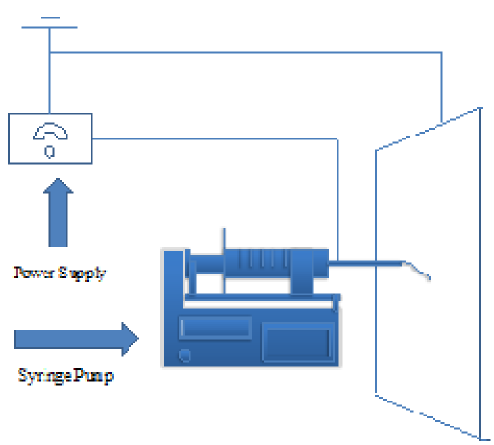

Figure 1: Experimental setup diagram. The syringe tip was connected to the power supply which delivered 10KV of volrage. Due to electrostatic attraction between the syringe tip and grounded metal plate, a rod was extruded.

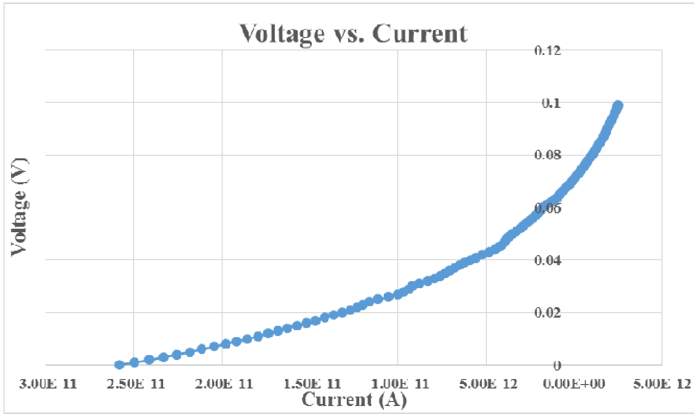

Figure 2: CHI 600E Electrochemical Analyzer supplied an increment voltage of 0.001 from 0 to 0.1 V.

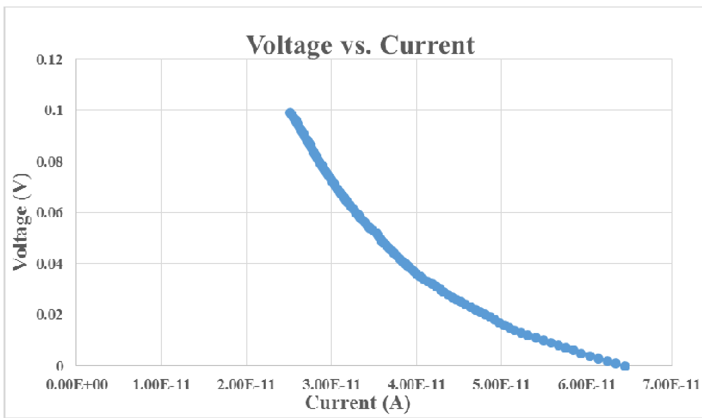

Figure 3: Same amount of voltage applied in a different test

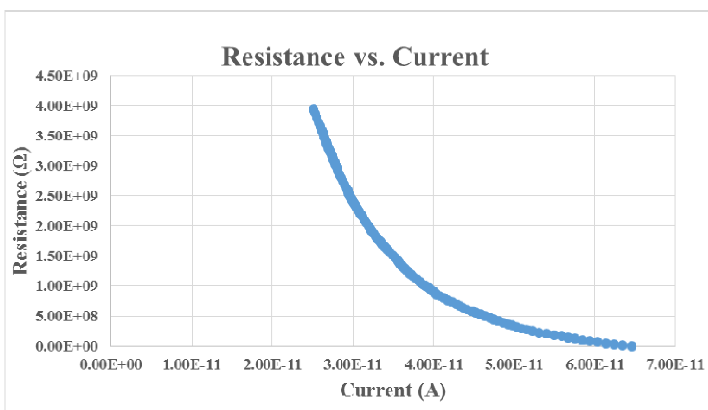

Figure 4: Referred to Figure 2. The graph presented the relationship between resistances vs. current

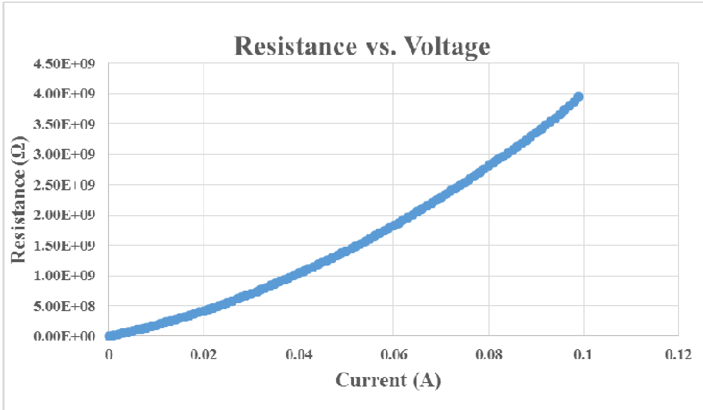

Figure 5: The graph presented the relationship between resistances vs. current

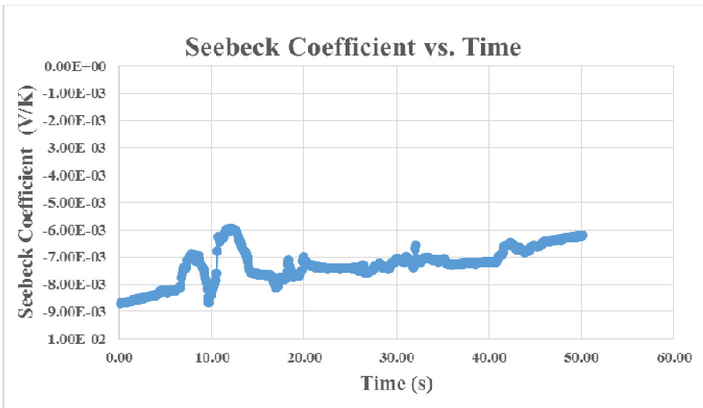

Figure 6: Seebeck Coefficient 319 Kelvin Temperature Gradient

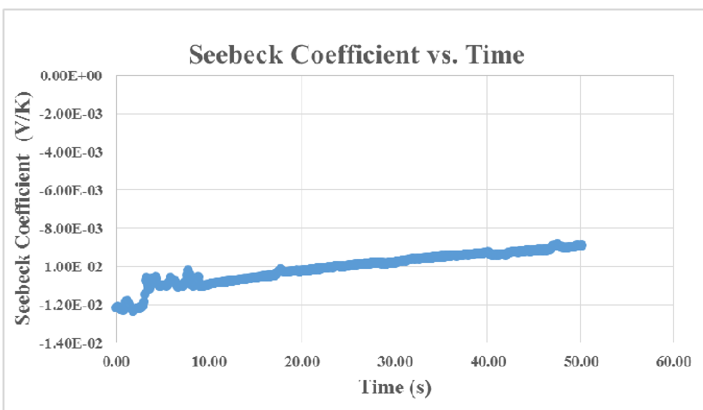

Figure 7: Seebeck Coefficient 321 Kelvin Temperature Gradient

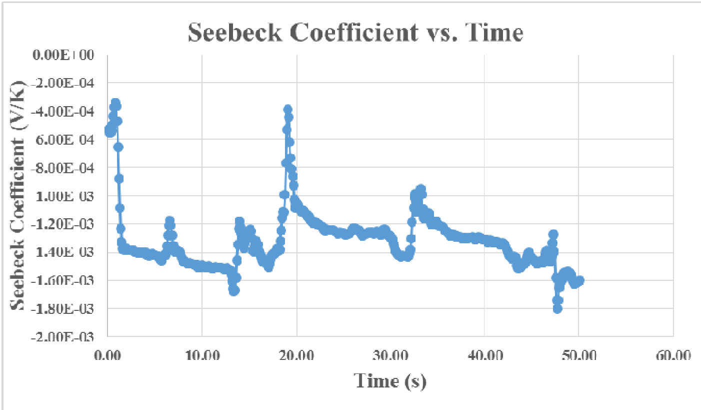

Figure 8: Seebeck Coefficient 325 Kelvin Temperature Gradient

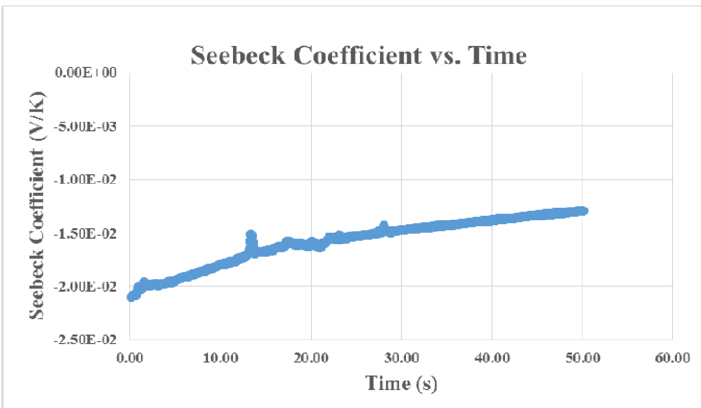

Figure 9: Seebeck Coefficient 331 Kelvin Temperature Gradient

CONCLUSION

In summary, this experiment investigates the thermoelectric property of lead telluride doped with silicone matrix under electrospinning. The material exhibits higher Seebeck coefficient and shows larger electrical resistance. This is due to the fact that Silicone rubber functions as an insulator. The mixture showed high Seebeck coefficient value. The results showed significant increase in Seebeck coefficient because of silicone rubber's hydrophobic ability to inhibit current leakage. The loss of hydrophobicity of silicone rubber will result in a conductive water film being form on the surface [24]. Leakage current is the inevitable loss of the current under high voltage. The other set of experiment tested electrical properties of this mixture. The highest electrical resistance measured was $5.93*10^{10}$ ohms. Hence, the highest electrical resistivity was calculated to be 9.31*105 ohm*m. Since majority of the energy in industries are lost as waste, Lead Telluride's intrinsic properties can offer promising potentials in the development of thermoelectric.

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