TEMPERATURE MEASUREMENTS USING A LITHIUM NIOBATE (LiNbO₃) PYROELECTRIC CERAMIC

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ABSTRACT

Temperature measurements from a power generation system could improve its overall safety and efficiency. In addition, real time temperature measurement could lead to a qualitative assessment of the health of the energy system. The challenge of real time temperature measurement inside of a power generation unit lies in the sustainability and the reliability of sensing materials in high temperature and corrosive environments. In this paper, a pyroelectric ceramic material (Lithium Niobate, LiNbO₃) capable of sustaining high temperatures, is considered as the temperature sensing material. LiNbO₃ is a pyroelectric material that generates current proportional to the rate of temperature change with time. This property is sustained by LiNbO₃ until it reaches its Curie temperature (1210 °C). The generated current could be used for many applications including temperature sensing and energy harvesting from waste heat recovery. This paper presents the temperature measurement results using a LiNbO₃ pyroelectric ceramic. Ceramics with dimensions of 1 cm x 1 cm with two different thicknesses, 2 mm and 1 mm, were used as the temperature measurement sensors. Two samples were heated by a 14.5 W/cm² heat flux and cooled with natural convection. Electrodes were made on both surfaces of the samples with high purity silver paint and two electrical leads were connected to a picoammeter to measure the generated current with respect to the rate of temperature change of the material during heating and natural cooling. A K-type surface thermocouple was attached on top of LiNbO₃ to compare the temperature measurements and maximum 8 % and 6.7 % differences were found for 2 mm and 1 mm thick samples, respectively.

KEYWORDS: Pyroelectricity, Sensor, Lithium Niobate, Temperature Measurement

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1. INTRODUCTION

Efficient operation of energy systems improves power output and reduces the harmful emission of pollutants, which include greenhouse gases such as CO₂ and NO[1]. Among the different processing parameters needed to accomplish this goal, an important parameter is the precise temperature monitoring of the system. Thermodynamics dictates that higher theoretical performance can be achieved by maintaining a higher or even optimum temperature throughout the energy conversion process[2]. However, materials used in current power generation units have some practical limits that can cause catastrophic failure of components if exposed to high temperatures[3-4]. Due to this constraint, a temperature safety margin is maintained for operating the system below the optimal temperature threshold to avoid thermally induced failure. If the operating temperature is not well characterized, then this safety margin becomes higher. However, this safety margin can be minimized by monitoring the temperature in real time during the combustion process. Nevertheless, continuous temperature control imposes a challenge in moving systems and corrosive environments. Hence, real time temperature monitoring is a major area of research due to its direct impact on thermal efficiencies and the structural integrity of these systems. Therefore, developing a temperature measurement method for a system operating at high temperature and harsh environment will be critical to run the system close to its optimal conditions. The most commonly used intrusive temperature measurement methods are thermocouples and resistance thermometry with a wide measurement range. However, these types of devices have a limited life cycle for particular applications where they are exposed to chemical reactions and erode due to long term exposure to the high temperature environment[5-6].

The work presented in this paper shows the measurement of temperature within the range between room temperature and 100 °C using the pyroelectric property of lithium niobate (LiNbO₃) as a sensing material. This type of material exhibits strong temperature variation dependent spontaneous polarization and loses its pyroelectric property when it is heated above its Curie temperature[7-8]. LiNbO₃ has a high Curie temperature (1210 °C) compared to any other pyroelectric ceramic materials hence it is promising for high temperature and harsh environment applications, which could be used for high temperature measurement systems.

Pyroelectric materials generate voltage and current when subjected to a change in temperature during heating and cooling[9]. The pyroelectric effect is different from the piezoelectric effect where mechanical stress is responsible for the polarization. Pyroelectricity is also different from thermoelectricity where the induced current is the result of temperature difference between hot and cold ends; this is known as the Seebeck Effect[10]. Pyroelectric current can be defined from the accumulated charge on the surface of the material, which is the change in charge per unit time[11]. Generated current (I) through a homogenous pyroelectric material with temperature T at any time (t) is presented in Eq. (1):

$$I = \frac{dQ}{dt} = -pA \frac{dT}{dt} \quad [1]$$

In Eq. (1) Q is generated charge due to temperature change, p is pyroelectric coefficient of the material, A is the surface area of the electrode and $\frac{dT}{dt}$ is the rate of temperature change of the material. By integrating Eq. (1) for a certain period of time the generated charge can be found, Eq. (2)[12]:

$$Q = \int_{t_i}^{t_f} I dt = \int_{t_i}^{t_f} -pA \frac{dT}{dt} dt = \int_{T_i}^{T_f} -pA dT = -pA(T_f - T_i) \quad [2]$$

Eq. (2) shows that the generated charge depends on temperature difference between the initial and final temperature instead of rate of temperature change. In this study a complete cycle of heating and cooling were applied to the sample material and the generated current from the sample was measured. The measured current due to the rate of temperature change was used to determine the temperature of the material. The focus of this study is to provide a measurable amount of current after applying a heating and cooling cycle to the LiNbO_3 and the temperature of the sample at any time (T_f) can be calculated from Eq. (2) by considering the initial temperature (T_i) as room temperature.

A previous study shows one method that can be used to transfer data wirelessly about the operating condition of a power generation unit using thermoelectric current from thermoelectric generator to a remote location[13]. Although pyroelectric current is different from thermoelectric current, it is possible to use LiNbO_3 as a pyroelectric current source instead of a thermoelectric generator in the system presented in Ref. [13]. Pyroelectric current can be used not only for temperature measurement sensors but also for waste energy harvesting. Any source of waste energy with temperature changes over time is suitable to produce pyroelectricity and can be harvested for many applications [12,14-17]

2. EXPERIMENTAL PROCEDURE

In this experiment the generated current output of the pyroelectric ceramic material LiNbO_3 due to heating and cooling was characterized. Small samples of LiNbO_3 (1 cm x 1 cm) with two different thicknesses (2 mm and 1 mm) were used for testing. Properties of the material are shown in Table 1.

Electrodes were made for the top and bottom surfaces of the sample by using high purity silver paint (SPI Supplies, CAS #: 7440-22-4, Structure Probe, Inc.). Two electrical leads were attached to the top and bottom surfaces of the sample with non-conductive paste. A silicon-heating pad

was used to apply a 14.5 W/cm^2 heat flux to the bottom of the sample for 210 s and then allowed to cool naturally for the following 300 s. An increasing rate of temperature change of the sample by silicon heating pad with 14.5 W/cm^2 heat flux was found for the first 210 s and then the rate of temperature change started to decrease and becomes near zero. Since the pyroelectric current is a function of rate of temperature change, during the heating cycle, the near zero rate of temperature change was not considered and then the cooling cycle started. The power supply to the heating pad was cut off after 210 s and the sample was allowed to cool naturally for the next 300 s and have a rate of temperature change with a negative slope and also have a near zero rate of temperature change at the end of cooling cycle. A K-type surface thermocouple was mounted on top of the sample, which utilizes the temperature of the sample surface as input for the temperature controller (Fig. 1). Under the same heating flux (14.5 W/cm^2); the sample with 2 mm thickness experienced a rate of temperature change as high as $0.5 \text{ }^\circ\text{C/s}$ and $0.3 \text{ }^\circ\text{C/s}$ during heating and cooling, respectively. The 1 mm thick sample experienced a rate of temperature change as high as $0.62 \text{ }^\circ\text{C/s}$ and $0.4 \text{ }^\circ\text{C/s}$ during heating and cooling, respectively. Two electrical leads from the sample were connected to an auto ranging picoammeter (KEITHLEY 485). The input current to the picoammeter forced through a feedback resistor inside of the picoammeter and inverted analog output voltage from the operational amplifier was connected to the data acquisition board. The analog output voltage from the picoammeter was measured with a LabVIEW VI set to a 500 Hz sample rate using a data acquisition board. Later the voltage readings were converted to a current value with a conversion factor provided by the picoammeter manufacturer. Power input to the silicon heating pad was controlled by a temperature controller and a solid state relay. The schematic diagram of the experimental setup is shown in Fig. 1.

The given temperature profile to the sample from a constant source of heat flux was measured by the surface thermocouple and the rate of temperature change was calculated from those measured value. Generated current from the pyroelectric material due to rate of temperature change was used to measure the temperature that the sample was experiencing and was compared with the sample temperature measured by thermocouple.

3. RESULTS AND DISCUSSION

Experimental results are shown here to determine the temperature from generated current of the pyroelectric ceramic material. The temperature profiles and rates of temperature change are shown for 2 mm and 1 mm thick samples in Figs. 2 and 3, respectively.

In Figs. 2 and 3 both the temperature profile and rate of temperature change show that the 1 mm thick sample reached a higher temperature with a higher rate of temperature change than the 2 mm thick sample for first 210 s heating. The principles of conduction heat transfer states

that for the same heat flow, cross sectional area, and material conductivity the temperature difference between the surface in contact with the heating pad and top of the LiNbO_3 must be lower for a thinner sample [18]. Therefore, the thermocouple mounted on top of the sample measures a higher temperature for the 1 mm thick sample compared to the 2 mm thick sample. Figs. 4 and 5 show the current response with the rate of temperature change for both sample thicknesses. From Eq. (1), the pyroelectric current (I) is a function of the pyroelectric coefficient (p) of the material, surface area of the electrode (A) and the rate of temperature change ($\frac{dT}{dt}$). The pyroelectric coefficient and the area of the electrode are the same for both samples. As the thinner sample experiences a higher rate of temperature change, the 1 mm sample responds with a higher amount of current output than the 2 mm thick sample.

Both current profiles in Figs. 4 and 5 matches with the rate of temperature change profile within experimental uncertainties satisfying Eq. (1). Figs. 6 and 7 show the current generation and corresponding temperature profile of thermocouple mounted on top of 2 mm and 1 mm thick sample, respectively.

Since LiNbO_3 could accurately measure the rate of temperature change, the feasibility of using LiNbO_3 instead of a thermocouple to sense temperatures in real time was then investigated. The temperature of the sample at any time can be calculated from Eq. (3), after rewriting Eq. (2) and considering initial temperature as room temperature.

$$T_f = -\frac{1}{pA} \int_{t_i}^{t_f} Idt + T_i \quad [3]$$

Here, T_i is the initial temperature, which is considered as room temperature at time t_i and T_f is the final temperature of the sample at time t_f . By identifying the initial temperature (T_i), pyroelectric coefficient (p), area of the electrode (A), and the total amount of current generated by the sample with the certain period of time (t_i to t_f), the final temperature of the sample (T_f) at any time can be calculated. To calculate the total amount of current, (area

under the current profile, $\int_{t_i}^{t_f} Idt$), numerical integration with Simpson's one-third rule was applied. The same process was applied to predict the temperature for both 2 mm and 1 mm thick LiNbO_3 samples. Figs. 8 and 9 show the results of measured temperature profiles by LiNbO_3 using their current output and Eq. (3).

Figs. 8 and 9 show the comparison between temperatures measured by thermocouple and LiNbO_3 . A maximum of 8 % and 6.7 % differences were found for the 2 mm and 1 mm thick sample, respectively. These differences are caused since the surface thermocouple is mounted on top of the sample. The temperatures measured by LiNbO_3 (dotted line in Figs. 8 and 9) were calculated from the generated current due to the rate of temperature change experienced by their contact with the silicon heating pad. On the other hand, the thermocouple is mounted on top of the LiNbO_3 and both the thickness and silver paint act as a thermal barrier. Also the top surface of the LiNbO_3 and the thermocouple is open to the atmosphere, which causes a heat loss due to natural convection resulting in lower measurements by the surface thermocouple than the LiNbO_3 is actually experiencing. Therefore, in Figs. 8 and 9, the differences between the temperatures measured by thermocouple and LiNbO_3 are observable where the dashed line represents the measurement by thermocouple and the dotted line represents the results from the LiNbO_3 is actually experiencing. Here, the 2 mm thick LiNbO_3 has a higher difference than the 1 mm thick sample as the thickness acts as a dominating factor in the thermocouple readings.

In our hypothesis, the top sample will experience a lower temperature than the bottom sample due to thermal resistance. To prove our hypothesis, two 1 mm thick LiNbO_3 with the same dimensions (1 cm x 1 cm) were stacked together by placing a thermocouple between them to create a sandwich structure. As shown in Fig. 10, this sandwiched structure sample is placed on top of the silicon heating pad and an experiment was conducted by maintaining all other parameters constant with respect to previous tests, including heat flux, heating, and cooling time. Measured temperature by the thermocouple was compared with those measured by both the bottom and top LiNbO_3 .

As shown in Fig. 10, the result of temperature measured from the output current of the bottom LiNbO_3 (dotted line) is higher than the measured temperature (dashed line) by the thermocouple as the thermocouple is measuring less temperature and the bottom sample is actually experiencing higher temperature. On the other hand, the temperature measured by the thermocouple is higher than the top LiNbO_3 is actually experiencing, resulting temperature measured by top LiNbO_3 (circled line) is lower than the temperature measured by thermocouple (dashed line). Also the bottom sample, thermocouple, and top sample reached their highest temperature after 213 s, 214 s and 219 s respectively, which imply the delay in measurement by the thermocouple and top sample. The result in Fig. 10 explains the reason for the differences in Figs. 8 and 9 between the temperature measurement by the thermocouple and LiNbO_3 . It is possible to minimize the difference by minimizing the heat loss and also by using thinner LiNbO_3 .

Based on results presented in this paper, it is feasible that LiNbO_3 ceramic materials could be used as a replacement sensor for low and high temperature applications. LiNbO_3 ceramic materials have several advantages over other invasive thermometry including thermocouples, resistive thermometry, and capacitive thermometry. For example, thermocouples require two homogeneous but dissimilar conductors requiring independent measurement of hot and cold junction temperature. The temperature sensor using LiNbO_3 is a single material and requires only room temperature as the reference point. Based on results presented in the paper, the accuracy and response rate is comparable to those from a conventional thermocouple. Any differences in the two measurements were due to convective heat losses.

4. SUMMARY AND CONCLUSIONS

Current responses of lithium niobate (LiNbO_3) material for two different thicknesses were experimentally measured by a picoammeter. The applied heat flux to the LiNbO_3 was 14.5 W/cm^2 and the generated pyroelectric current was the result of the rate of temperature change of the sample. This generated current was then used to calculate the temperature of the LiNbO_3 . Numerical integration with Simpson's one-third rule was applied to the pyroelectric current profile to calculate the total amount of current for a certain period of time. The integration results were used to measure the temperature of the LiNbO_3 at any time and compared with the temperature measured by thermocouple mounted on top of it. Maximum differences of 8% and 6.7% were found between the temperature measurement by the thermocouple and LiNbO_3 for 2 mm and 1 mm thick samples, respectively. The difference between temperature measurements is due to the lower temperature measurement by the surface thermocouple, which is explained in an experiment with a sandwich structure sample. The difference observed can be minimized by maintaining a low amount of heat loss from the sample and also using a thinner sample. Overall, the results from this study show a methodology of temperature measurement using the pyroelectric properties of lithium niobate ceramic material between room temperature and 100°C . Measurement at high temperature using lithium niobate could be possible as it can withstand high temperature and has a stable pyroelectric property until the material reaches its Curie temperature of 1210°C . This study is thought to be of interest to those designing and developing similar type of temperature sensors for various industrial and commercial applications.

Nomenclature

I	Pyroelectric Current	(A)
Q	Pyroelectric Charge	(C)
p	Pyroelectric Coefficient	(C/°C·cm ²)
A	Area of the sample (cm ²)	
T_i	Initial Temperature of the sample [Room Temperature]	(°C)
T_f	Final Temperature at any time	(°C)
t_i	Initial Time	(s)
t_f	Final Time	(s)
dT	Change of Temperature of the sample	(°C)
dt	Change of Time	(s)

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Table 1: Properties of lithium niobate, LiNbO_3 (Gooch & Housego Palo Alto)

Dimension (LiNbO_3)	1×1 cm, 2 mm and 1 mm in thickness
Density	4.65 g/cm ³
Curie Temperature	1142.3 ± 0.7 °C
Relative Dielectric Constant @ 25 °C (unclamped <500 KHz)	$\epsilon_{11} = 85$, $\epsilon_{33} = 28.7$
Pyroelectric Coefficient (@ 25 °C)	-8.3×10 ⁻⁵ C/°C·cm ²
Piezoelectric Strain Coefficients (@25°C ×10 ⁻¹² C/N)	$d_{12}= 69.2$, $d_{31}=0.85$, $d_{22}=20.8$ $d_{33}=6.0$
Thermal Conductivity (@ 25 °C)	10 ⁻² cal/cm·s·°C

Figure 1: Schematic diagram of the experimental setup

Figure 2: Temperature profile and rate of temperature change of the thermocouple mounted on top of the 2 mm thick LiNbO₃

Figure 3: Temperature profile and rate of temperature change of the thermocouple mounted on top of the 1 mm thick LiNbO₃

Figure 4: Generated current by LiNbO_3 and rate of temperature change of thermocouple mounted on top of 2 mm thick LiNbO_3

Figure 5: Generated current by LiNbO_3 and rate of temperature change of thermocouple mounted on top of 1 mm thick LiNbO_3

Figure 6: Generated current by LiNbO_3 and temperature profile of thermocouple mounted on top of the 2 mm thick LiNbO_3

Figure 7: Generated current by LiNbO_3 and temperature profile of thermocouple mounted on top of the 1 mm thick LiNbO_3

Figure 8: Temperature measured by thermocouple and 2 mm thick LiNbO₃

Figure 9: Temperature measured by thermocouple and 1 mm thick LiNbO₃

Figure 10: Temperature measured by the thermocouple and 1 mm thick sandwich structured LiNbO_3