Performance and Characterization of Seebeck Coefficient and Power Factor in CMC/Glycerin Gel Electrolyte Based Ionic Thermoelectric

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Abstract *- Ionic thermoelectric (i-TE) materials have gained significant attention for their potential to convert low-temperature thermal energy into electrical energy. In this study, gel electrolyte-based i-TE materials have been synthesized using carboxymethyl cellulose (CMC), glycerin and* H_3PO_4 *solution as electrolyte. Analysis of thermoelectric properties such as Seebeck coefficient and power factor of the gel electrolyte has been carried out. In this study, ionic conductivity and potential difference or output voltage of CMC/glycerin gel electrolyte were measured. The results of this study indicate that the ionic conductivity and output voltage of the CMC/glycerin sample increase with temperature, whereas the Seebeck coefficient and power factor tend to decrease as the temperature gradient between the hot and cold parts of the CMC/glycerin gel electrolyte sample increases. These findings suggest that the CMC/glycerin-based polymer gel electrolyte has potential for use in i-TE devices, particularly in applications where high-power output is required.*

Keywords*: Ionic Thermoelectric; Gel Electrolytes; Seebeck Coefficient; Power Factor; CMC*

INTRODUCTION

Solar irradiation on surfaces such as building roofs, concrete structures, and photovoltaic cells can generate thermal energy. This presents challenges in environmental and energy systems management. This thermal energy, usually has a relatively low temperature, ranging from 40°C to 60°C. This thermal energy is often not utilized and simply released into the surrounding environment (Serale et al., 2014). Thermoelectric (TE) generators are one of the promising tools to capture and convert this thermal energy into electric power. The energy conversion mechanism in a thermoelectric generator is the conversion of thermal energy into electrical energy through the movement of charge carriers in response to a temperature gradient. This phenomenon is called the Seebeck effect. Traditional TE generators mostly use semiconductor-based materials (s-TE) (Zhang & Zhao, 2015) (Osorio et al., 2023). Based on research, the Seebeck coefficient value of s-TE can reach 561 μV/K on a thin layer of PbSe nanocrystals (Sousa et al., 2022). Although s-TE materials are well suited for high temperature applications (Xie & Gupta, 2020), their application at lower temperatures is hindered by reduced efficiency and increased thermal conductivity.

Ionic thermoelectric (i-TE) materials are being developed as alternative materials for TE generators. i-TE materials use ions such as cations and anions as charge carriers to create electric voltage. This mechanism is different from s-TE materials that use electrons and holes as charge carriers. The ionic thermoelectric mechanism is able to increase the Seebeck coefficient and decrease the thermal conductance. Thereby increasing the efficiency of heat-to-electricity conversion at relatively low temperatures. (Wu et al., 2021). One study revealed the seebeck coefficient of i-TE materials to be 32.7 mV/K, which is much larger than that of s-TE substances (He et al., 2023). In addition, i-TE materials are more affordable and have limited environmental pollution than s-TE materials (Zhao et al., 2021). One of the i-TE materials is polymer electrolyte. They have shown promising results in applications such as thermoelectric generators and energy storage devices (Muddasar et al., 2024).

Carboxymethyl cellulose (CMC) is one of the materials that can be used for polymer gel electrolytes. CMC-based gel electrolytes exhibit high ionic conductivity. These materials exhibit relatively low thermal conductivity when compared to other materials such as semiconductors. One study found that the thermal conductivity of CMC gel electrolytes is about 0.1 W/mK. While another study reported the thermal conductivity of this material is 0.2 W/mK (Akhlaq et al., 2023). Low thermal conductivity values are required in TE generators to obtain higher output performance (Bhuiyan et al., 2022). The ionic conductivity of gelled polymer electrolytes can be improved by adding plastizers such as glycerin. High ionic conductivity can improve the performance of i-TE materials (Ali et al., 2022). Based on the advantages of electrical and thermal conductivity, glycerin-supplemented CMC gel electrolytes have the potential to be applied as i-TE materials.

However, there is no information from research results or scientific articles related to the performance and characteristics of thermoelectricity in CMC/glycerin gel electrolyte at relatively low temperatures, especially below 70 ^{o}C. The TE performance and characteristics include output voltage, Seebeck coefficient and power factor. The CMC/glycerin gel electrolyte needs testing to verify its performance and characteristics. Therefore, in this study, the synthesis and testing of thermoelectric performance and characteristics on CMC/glycerin gel electrolyte were carried out. The test was carried out at a temperature variation below 70 ^{o}C. The main objective is to analyze the thermoelectric characteristics and performance of CMC/glycerin gel electrolyte. The thermoelectric characteristics and performance analyzed are ionic conductivity, output voltage, Seebeck coefficient and power factor. Seebeck coefficient, power factor, and temperature correlation are crucial for efficient i-TE generators converting low-temperature heat to electricity.

RESEARCH METHODS

a) Raw Materials

The raw materials used for the polymer gel electrolyte were CMC and glycerin. CMC is a polymer that is easily soluble in water and can be doped with electrolyte solutions, thereby enabling high ionic conductivity. Glycerin is a polymer plasticizer that can increase ionic mobility in polymers. The electrolyte used for the gel electrolyte was H_3PO_4 solution with a concentration of 85%. H_3PO_4 has been widely used in research as an electrolyte in polymer electrolyte materials. addition of H_3PO_4 in CMC polymer gel enhances proton conduction. The solvent used to dissolve CMC was distilled water. All raw materials were obtained from local chemical stores.

b) Gel Electrolyte Synthesis

CMC/glycerin gel electrolyte samples were synthesized with the following steps: CMC gel preparation, mixing with glycerin and H_3PO_4, and gel solidification process. CMC gel was made by mixing 1 g CMC powder with 24 g distilled water and stirring until a homogeneous gel was formed. All raw materials are weighed using a digital analytical scale. At the mixing stage, 4 g glycerin and 5 g H_3PO_4 solution were added to the CMC gel, then stirred until a homogeneous gel electrolyte was formed. The mixing and stirring process for making gel electrolyte is carried out in a clean beaker. In the molding stage, 50 g of mixed CMC/glycerin gel electrolyte was placed in a clean petri dish. The gel electrolyte was left in a closed container at room temperature for 10 days to enhance its solidification. The closed container is intended to protect the gel electrolyte from contamination by dirt or dust.

c) Ionic Conductivity Measurement

Ionic conductivity measurement using a series of devices referring to Figure 1. CMC/glycerin gel electrolyte samples were put into a 4 mm inner diameter glass pipe. The gel electrolyte sample is sandwiched until there is no empty space using a pair of copper piston. Copper piston is solid cylinder of copper with a diameter of 4 mm. The copper piston was serves as a sample holder and electrode to conduct electric current to the sample.

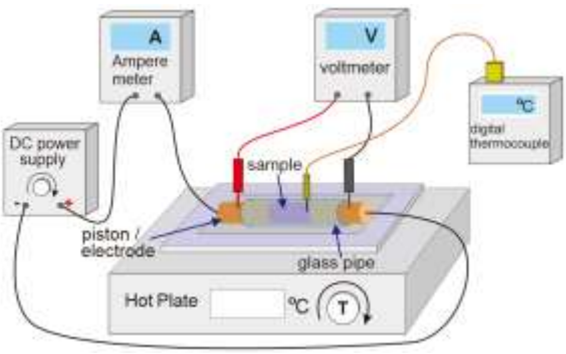

Figure 1. Ionic conductivity mesurement setup

The input current in this measurement was varied with 3 different values, so that 3 different voltage were obtained on the sample. Current (I) and voltage (V) values were measured using a sanwa CD800a type multimeter. Ionic conductivity measurements on polymer gel electrolyte samples were carried out at temperatures 30, 35, 40, 45, 50, 55, 60 and 65 °C. The value of electrical conductivity (σ) could be calculated using equation 1.

$$\sigma = \frac{d}{R_B A} \tag{1}$$

Based on equation 1, d is the length of the sample in the glass cylinder (m). R_B is the electrical resistance of the sample (ohm). A is the cross-sectional area of the sample in the glass cylinder (m^2). R_B value is calculated from the slope of the linear regression of the voltage vs current relationship graph (I-V graph). The slope value of the linear regression can be calculated by equation 2.

$$R_B = \frac{n\sum_{i=1}^{n} x_i y_i - \sum_{i=1}^{n} x_i \sum_{i=1}^{n} y_i}{n\sum_{i=1}^{n} x_i^2 - (\sum_{i=1}^{n} x_i)^2} \tag{2}$$

From equation 2, n is the number of measurements. x_i is the data of the current passing through the sample. y_i voltage data on the sample when the current passes through the sample. The use of linear regression is intended to eliminate bias in resistance values caused by contact resistance between the electrode and the sample and errors in reading values on voltage and current measuring instruments.

d) Potential Difference Measurement

Devices arrangement for measuring the potential difference or output voltage is shown in Figure 2. The gel electrolyte samples were molded on plastic pipes with a size of 4 mm inner diameter, 5 mm length and 1 mm thick pipe skin. This measurement was carried out at several temperature 30, 35, 40, 45, 50, 55, 60 and 65 °C. The sample was heated using a digital hot plate. The voltage was measured digital multimeter. The temperature was measured with digital thermocouple.

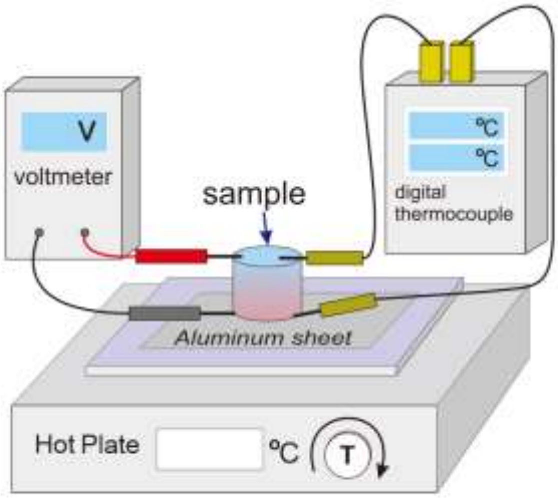

Figure 2. Difference potential measurement setup

e) Analysis of Seebeck Coefficient and Power Factor

The value of the Seebeck coefficient was calculated using equation 3. Based on equation 3, S is the seebeck coefficient (V/K). ΔV is the potential difference of the sample when heated (V). ΔT is temperature difference between the hot part and the cold part of the sample (K) (Yang & Portale, 2021).

$$S = \frac{\Delta V}{\Delta T} \qquad (3)$$

In this study, the power factor was calculated using equation 4. PF is the power factor value (W/mK^2). σ is the ionic conductivity value of the electrolyte gel sample. S is the Seebeck coefficient (Li et al., 2022).

$$PF = \sigma S^2 \qquad (4)$$

f) Calculation of Activation Energy

The activation energy was calculated using the gradient value of the Arrhenius conductivity graph. The relationship of the Arrheniuss curve slope to the activation energy is shown by equation 5.

$$b = -\frac{E_a}{k_B} \qquad (5)$$

E_a is the activation energy, k_B is the Boltzmann constant and b is Arrhenius curve slope. The Arrheniuss conductivity curve refers to Arrhenius conductivity equation shown by equation 6 (Johannes et al., 2022) (Rathod et al., 2015).

$$\sigma_T = \sigma_o \exp\left(-\frac{E_a}{k_B T}\right) \qquad (6)$$

σ_{Ti} is ionic conductivity value at temperature T_i (mS/m). σ_o is pre-exponential factor. T_i is the temperature in Kelvin.

RESULTS AND DISCUSSION

Result

a) Gel Electrolyte

The electrolyte gel synthesized from CMC, glycerin and H_3PO_4 solution in this study is shown in Figure 3. In this study, the synthesized gel electrolyte is transparent, and there are many air bubbles. The air bubbles contained in the gel electrolyte are caused by the stirring process when mixing CMC gel with glycerin.

Figure 3. CMC/glycerine gel electrolyte

b) Ionic Conductivity

The results of ionic conductivity measurements of CMC/Glycerin gel samples are shown in Table 1.

Table 1. Table of ionic conductivity

No.	Temperature (ºC)	Ionic Conductivity (S/m)
1.	30	0,244
2.	35	0,251
3.	40	0,356

No.	Temperature (°C)	Ionic Conductivity (S/m)
4.	45	0,401
5.	50	0,445
6.	55	0,684
7.	60	0,855
8.	65	1,09

Based on Table 1, the increase in temperature from 30 to 65 °C is followed by an increase in electrical conductivity. The increase in conductivity tends to be non-linear to the increase in temperature.

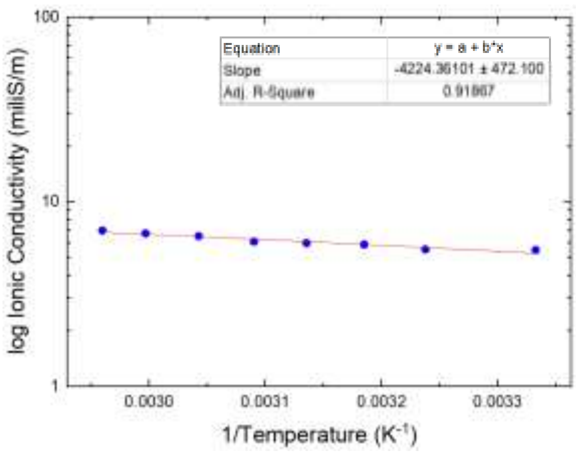

Figure 4. Ionic conductivity at varying temperature in Arrhenius plot

Figure 4 shows the relationship between electrical conductivity and temperature of CMC/glycerin gel polymer in Arrhenius graph.

c) *Potential Difference*

The results of the potential difference measurement on the CMC/glycerin gel electrolyte sample are shown in Figure 5.

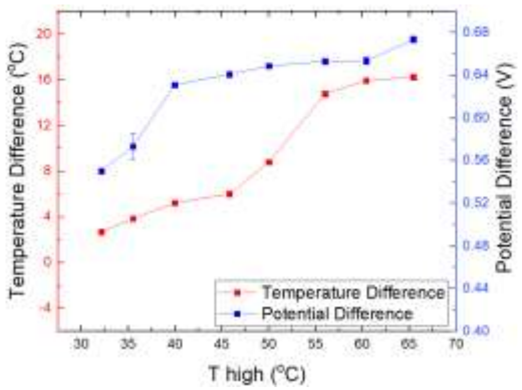

Figure 5. Temperature and potential difference at varying temperature

Figure 5 shows an increase in heating temperature (T high) followed by an increase in potential difference (*ΔV*) or output voltage of CMC/glycerin gel electrolyte samples. The highest output voltage is 0.673 V at 65 °C, while the lowest output voltage is 0.55 V at 30 °C. The graph in Figure 5 shows that the temperature difference (*ΔT*) in the sample increases as the temperature (T high) increases. The sample temperature difference is the temperature difference in the hot part (T high) with the temperature of the cooler part (T low) in the CMC/glycerin gel electrolyte sample.

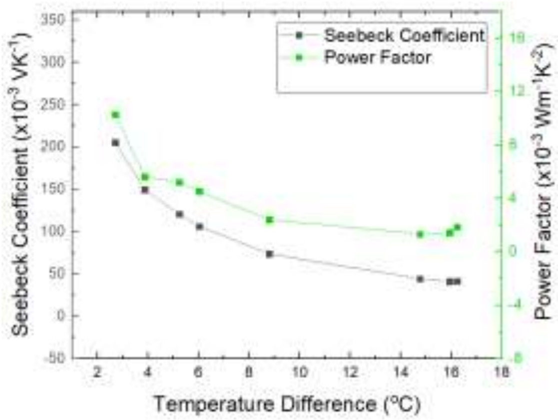

Figure 6. Seebeck coefficien and power factor at varying temperature.

d) *Seebeck Coefficient and Power Factor*

The results of the calculation of the Seebeck coefficient and power factor values on the CMC/glycerin gel electrolyte sample at varying temperature are shown in Figure 6. The highest Seebeck coefficient is 0.206 V/K at 30 °C. The lowest Seebeck coefficient is 0.041 V/K at 65 °C. The highest power factor value is $1{,}03 \times 10^{-2}$ W/mK^2 at 30 °C, and the lowest is 1.87×10^{-3} W/mK^2 at 65 °C.

The relationship between Seebeck coefficient, power factor and potential difference of CMC/glycerin gel electrolyte sample is shown in Figure 7.

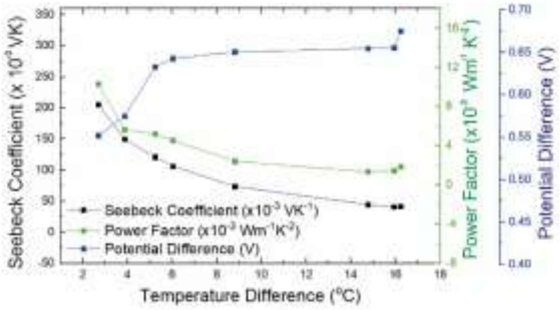

Figure 7. Seebeck coefficient, power factor and potensial difference at varying temperature difference.

Discussion

The electrical conductivity of the gel polymer electrolyte is strongly influenced by the mobility of ions in the gel polymer matrix. Ion mobility determines the value of ionic conductivity in the gel polymer electrolyte. The electrical conductivity of the material can be represented by ionic conductivity. Based on the conductivity test results, the electrical conductivity of the CMC/glycerin gel electrolyte increases with increasing temperature. This result is in accordance with the ionic conductivity value of the results of research conducted by Johannes and the team on Polyurethane-LiTFSi based polymer electrolyte materials published in the 2022 article. The results showed that increasing temperature can increase ionic conductivity in polymer electrolytes (Johannes et al., 2022). This result is in accordance with the theory or mechanism of the relationship between ionic conductivity and temperature in polymers. increasing temperature will increase ionic conductivity. this is due to increased ion mobility due to increased heat in the polymer electrolyte.

Increasing temperature can cause ions from the H_3PO_4 electrolyte to move more easily or have higher mobility in the CMC/glycerin gel polymer matrix. Higher temperatures cause higher polymer chain vibrations so that the free volume in the gel polymer tends to enlarge. A large free volume can result in easier ion transfer in the polymer gel (Johannes et al., 2022) (Gupta et al., 2021). An increase in thermal energy can increase the kinetic energy of ions, thus the ions easily diffuse in the gel electrolyte matrix (Aziz et al., 2018) and improve ion transfer in the gel polymer matrix (Shao et al., 2020).

Based on Figure 4, the activation energy can be calculated from the slope of the Arrhenius graph. The activation energy value of ions in the CMC/glycerin gel electrolyte sample is 0,364 eV. Activation energy is the energy required by ions to be able to move in the gel polymer matrix.

Based on Figure 5, the potential difference of the CMC/Glycerin Gel electrolyte exhibits a positive correlation with temperature, demonstrating an upward trend as the temperature rises. This result is in accordance with the Seebeck effect mechanism, the greater the temperature difference between the hot and cold parts of the TE material, the greater the potential difference in the material.

The generation of the potential difference due to the application of heat to the sample can be explained by the Soret effect. The Soret effect is the movement or migration of atoms or ions in a camporan material (such as a polymer electrolyte) caused by a temperature gradient in the material. The temperature gradient causes cations and anions to separate resulting in charge polarization in the gel electrolyte (Cheng & Ouyang, 2022) Anions tend to be concentrated in the hot part while cations are concentrated in the cold part (Muddasar et al., 2024).

In the results of this study, the hot part of the polymer gel electrolyte sample has a positive pole and a negative pole in the cold part of the sample. Based on the soret effect, the temperature difference or gradient in the sample can cause the ions of the H_3PO_4

electrolyte contained in the CMC/glycerin gel electrolyte sample to be polarized. Cations (H^+) are concentrated in the cooler part, while anions (PO_4^{3-}) are concentrated in the hot part. This difference in ion concentration causes a difference in electrical potential between the hot and cold parts of the polymer gel electrolyte sample. The higher the temperature difference, the greater the difference in the concentration of cations and anions of H_3PO_4 electrolyte in the sample. this can increase the potential difference in the gel electrolyte sample (Chang et al., 2016).

The Seebeck effect, also called the thermoelectric effect, is a phenomenon that explains the generation of an electric potential difference when there is a temperature difference in thermoelectric materials. In this study, the Seebeck coefficient of CMC/glycerin gel electrolyte samples tends to decrease as the temperature increases. The change in seebeck coefficient value is shown in Figure 6.

The temperature dependent Seebeck coefficient is shown in Figure 6. As the temperature (T high) increases from 30 ^oC to 65 ^oC, the Seebeck coefficient value decreases. This decrease is attributed to the increasing temperature difference (ΔT) as the temperature (T high) increases. In addition, the increase in potential difference (ΔV) of the sample is not large as the heating temperature (T high) increases. This causes the ratio between the potential difference (ΔV) and the sample temperature difference (ΔT) to get smaller as the heating temperature (T high) value increases. The ratio of the potential difference value to the sample temperature difference is the Seebeck coefficient, this relationship is mathematically shown by the equation 3.

The power factor is determined by the Seebeck coefficient and the electrical conductivity of the thermoelectric sample. The power factor value indicates the efficiency level of the thermoelectric material. The relationship of power factor with electrical conductivity and Seebeck coefficient is shown in equation 4. In this study, the power factor value of CMC/glycerin gel electrolyte decreased as the temperature (T high) increased. The decrease in power factor value tends to follow the pattern of changes in the Seebeck coefficient as shown in Figure 6.

Figure 7. show the increase in temperature difference of CMC/glycerin gel electrolyte sample is followed by a decrease in the value of Seebeck coefficient and power factor. but the value of potential difference tends to increase as the temperature difference increases in CMC/glycerin gel electrolyte sample.

Based on the value of Seebeck coefficient and power factor in this study. CMC/glycerin gel electrolyte has a higher Seebeck coefficient and power factor than the results of previous research using liginin-based hydrogel material with 6M NaOH as an electrolyte. The material has a maximum Seebeck coefficient of 0.013 V/K and a maximum power factor of 3.38×10^{-3} W/mK^2 (Muddasar et al., 2024). While in this study, the CMC/glycerin gel electrolyte has a Seebeck coefficient value of 0.206 V/K and a power factor of 1.03×10^{-2} W/mK^2. Ionic thermoelectric materials based on CMC/glycerin gel electrolyte are very potential to be applied to thermoelectric generators (TEG) to convert low temperature thermal energy into electricity. An example of low temperature thermal energy is the thermal energy generated on the surface of objects such as roofs and concrete exposed to solar radiation.

CONCLUSION

The potential difference generated in the CMC/glycerin gel electrolyte tends to increase as the temperature of the material increases. A fairly high increase in potential difference occurs at temperatures of 30 to 40 ^{o}C. While the increase in potential difference at temperatures of 40 to 65 ^{o}C tends not to be high. The highest output voltage at 65 ^{o}C with a value of 0.67 V. The value of the Seebeck coefficient and power factor in this material decreases while increasing temperature in the sample. The highest Seebeck coefficient value is 206 mV/K at 30 ^{o}C, and the lowest is 41 mV/K at 65 ^{o}C. The highest power factor value is $1{,}03\times10^{-2}$ W/mK^2 at 30 ^{o}C, and the lowest is 1.87×10^{-3} W/mK^2 at 65 ^{o}C. Increasing temperature causes an increase in the value of ionic conductivity in CMC/glycerin gel electrolyte. The ionic conductivity increases from 0.244 S/m to 1.09 S/m in the temperature range of 30 to 65 ^{o}C. Based on the output voltage, Seebeck coefficient and power factor in this study, CMC/glycerin gel electrolyte can be applied for thermoelectric generator based on i-TE material. Future research are expected to explore other electrolyte combinations, analyze thermal conductivity values and figure of merit so that more comprehensive information is obtained regarding the performance and characteristics of the CMC/gel electrolyte.

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