

THE DIELECTRIC AND THERMOELECTRIC PROPERTIES OF 30Li₂O: 3MoO₃: 40Bi₂O₃: 20TeO₂: 7CuO GLASS CERAMIC MATERIAL

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Abstract

In this study, the physical and thermoelectric characteristics of glass ceramic materials are examined to provide the thermoelectric performance of 30Li₂O: 3MoO₃: 40Bi₂O₃: 20TeO₂: 7CuO samples that were synthesized utilizing the melt-quenching process. Archimedes' Principle was used to determine the physical properties of the sample, showing a density of 4.53 g/cm³. An X-ray diffractometer (XRD) used to analyze the crystal structure revealed the Bismuth Oxide phase. The results of measuring with the LCR Meter for the frequency range were set from 50 Hz to 1 MHz at room temperature, showing the dielectric constant (ϵ') and the dielectric loss (ϵ'') that tended to decrease with increasing frequency. Thermoelectric properties were examined in a temperature range of 300 K to 600 K indicating the electrical resistivity, the Seebeck coefficient, and the power factor. By calculating the efficiency of thermoelectric materials, the power factor term is considered at which temperature it shows its highest value. In this work, it was shown on 6.99 mW/mK² at a temperature of 474 K. Since this glass ceramic has a high degree of stability and efficiency, It has the potential to be used in thermoelectric devices.

Keywords: Dielectric constant; Melt-quenching; Power factor; Seebeck coefficient

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Introduction

Thermoelectric materials are another outcome of ongoing development activities in the energy sector, spanning the past and present. Thermoelectric materials can use the Peltier effect to create coolness and the Seebeck effect to convert heat energy from a source into electrical power. Thermoelectric devices are inexpensive because they do not have any moving components and operate silently. Because thermoelectric materials are tiny and do not include greenhouse effect components, compact design is made feasible, opening up a wider variety of applications. Terasaki *et al.* produced NaCo₂O₄, a single-crystal thermoelectric material, to demonstrate its efficiency (Terasaki, *et al.*, 1997). Other thermoelectric oxide materials may be found, such as SrTiO₃ (Ohta *et al.*, 2005), ZnO (Ohtaki *et al.*, 1996), and Ca₃Co₄O₉ (Shikano and Funahashi, 2003). These materials are known to be promising for high-temperature thermoelectric applications because they are chemically and thermally stable in air at high temperatures. The formula for calculating a thermoelectric material's performance is $ZT = S^2 \sigma T / \kappa$, where T is the absolute temperature, S is the Seebeck coefficient, and σ and κ are the material's electrical and thermal conductivity, respectively the difference in temperature between hot and cool junctions determined by ΔT . The qualities of the component materials determine the maximum value of accessible sources of heat and cold (thermal and chemical stability). To maximize ZT , minimize the κ (thermal conductivity) denominator or increase the numerator, $S^2 \sigma$ (power factor). In conclusion, strong and poor electrical conductivity is necessary for an effective thermoelectric material. Glass ceramics have high thermomechanical characteristics and are inexpensive, making them very valuable for many applications. They have many advantages, such as pore-free and very homogenous materials, large pieces, and various forms are possible using glassmaking processes, and significant transparent or translucent components can be made (Gonon and Dupla, 2021). Low gap semiconductors must be studied to ascertain whether glass ceramics exhibit thermoelectric characteristics. The literature reports on several glasses having semiconducting characteristics that contain heavy elements. Most of these glasses are based on pnictides, chalcogenides, and oxide glasses with significant concentrations of transition metal oxides (Petersen *et al.*, 1973; Varshneya and Mauro, 2019). TeO₂ based glasses have garnered much interest because of their excellent thermal stability ($\Delta T > 373$ K) (El-Mallawany, 1995; El-Mallawany and El-Mallawany, 2001;

Souri, 2017; Souri and Shahmoradi, 2017), low melting point (El-Mallawany, 1995; El-Mallawany and El-Mallawany, 2001; Dimitrov and Komatsu, 2013; Souri and Shahmoradi, 2017), and electronic polarizability values (Dimitrov and Komatsu, 2013; Souri, 2017). Chowdari *et al.* reported on the variation of Cu₂O and MoO₃ content in TeO₂ glass to study the glass structure and its relationship with thermal and electrical properties (Chowdari, 1998). The electrical conductivity of composite glasses made by the traditional melt quench technique (65-x)Bi₂O₃: xLi₂O: 20ZnO: 15B₂O₃ was investigated by Shashidhar and Syed, Under the frequency range of 100 Hz to 1 MHz, the AC conductivity was examined at temperatures between 30°C and 300°C (Bale and Rahman, 2012). The structural, dielectric, and electrical characteristics of the glass ceramic material 30Li₂O: 2MoO₃: 40Bi₂O₃: 20TeO₂: 7CuO are reported in this work to help researchers and interested parties learn more about thermoelectric materials and how to develop future thermoelectric properties and applications for the device.

Materials and Methods

Glass ceramic samples in the composition of 30Li₂O: 3MoO₃: 40Bi₂O₃: 20TeO₂: 7CuO were prepared by the traditional melt quenching technique with the purity of all chemical powders showing $\geq 99.99\%$. The chemical powders were melted in a porcelain crucible at 1,473 K for one hour in a consistent temperature zone of an electrically powered furnace. Onto a graphite platform, the molten liquid was poured into an area between two blocks of graphite rectangles at room temperature. The annealing procedure, which involves heating the sample to 473 K for two hours in the furnace, decreases stress and strain on the piece.



Figure 1. The glass ceramic samples were prepared to investigate the characterization

They were separated into three parts to characterize the samples and polished to different sizes. To facilitate an X-ray diffraction (XRD, SmartLab SE, Rigaku, Japan, Cu $K\alpha$ radiation) structural investigation, part of the material was ground into a powder. In Figure 1, the samples were produced as disks with a thickness of 1.0 mm to determine the dielectric characteristics via an LCR Meter (4284A Precision, Keysight, USA) with a frequency range of 20 Hz - 1 MHz. (iii) A sample with dimensions of approximately $2.0 \times 5.0 \times 10 \text{ mm}^3$ was prepared to determine the electrical characteristics via the Seebeck Coefficient/Electric resistance measurement system (ZEM-3 series, ULVAC, Inc., Japan) in a temperature range of 300 K to 600 K.

Results and Discussion

The powder was ground, and XRD was used to speed the analysis in a 2θ range of 10-80 degrees to ascertain the sample's structure. Figure 2, shows the X-ray diffraction spectra for the glass ceramic sample. In space group 225: Fm-3m, the diffraction peaks along [1 1 1], [2 0 0], [2 2 0], [3 1 1], [2 2 2], [3 3 1], and [4 2 0] correspond to the phase of Bi_2O_3 . (DB Card No. 38436). The broad on the left side of the XRD pattern indicates the presence of an amorphous phase within the glass structure, which is common in glass ceramic materials. The two superposed peaks of [1 1 0] and [2 0 0] suggest the existence of crystalline phases within the amorphous matrix. The amorphous part results from the rapid cooling during the melt-quenching process, which prevents the formation of long-range order. The crystalline atomic planes are due to the partial crystallization of the Bi_2O_3 phase, which is indicated by the distinct peaks in the XRD pattern. This cohabitation of amorphous and crystalline phases can enhance the material's properties by combining the flexibility of the amorphous phase with the stability and specific characteristics of the crystalline phase. Using water as the immersion liquid and the Archimedes principle, the glass sample's density (D) and molar volume (V_m) were measured at room temperature shown in Table 1. For AC measurements, the dielectric constant (ϵ') and the dielectric loss (ϵ'') were reported in Figures 3(a) and 3(b), respectively. Because of the Bi_2O_3 in the sample, Figure 3(b) shows that the dielectric loss (ϵ'') decreases with increasing frequency. The phase difference between the electric field that is applied and the polarization is the cause of this phenomenon. This glass ceramic sample has imperfections within its structure identified by X-ray diffraction as Bi_2O_3 . It exists in two oxidation

states, Bi^{3+} operates as a network modifier while Bi^{5+} acts as the original network. The Bi^{3+} is distinguished by the presence of a $6s^2$ lone pair of electrons, which causes the cation network to be highly polarizable (Sammes *et al.*, 1999; Zhao *et al.*, 2013). In Table 2, the electrical resistivity, the Seebeck coefficient, and the power factor of a glass ceramic material are shown as the electrical characteristics. On the concept of electrical conductivity in non-single-crystal solids. To explain the electrical conductivity of glass ceramic materials, it is more related to the density of states ($N(E)$) than band theory due to the distortion within the non-crystalline structure. As a result, the Seebeck coefficient, which ascertains the carrier concentration and the charge carrier as a function of the experiment's temperature, is represented by the Mott relation (Liu *et al.*, 2012). In the temperature measurement 474 K, the highest value of the Seebeck coefficient was shown as -0.09 V/K.

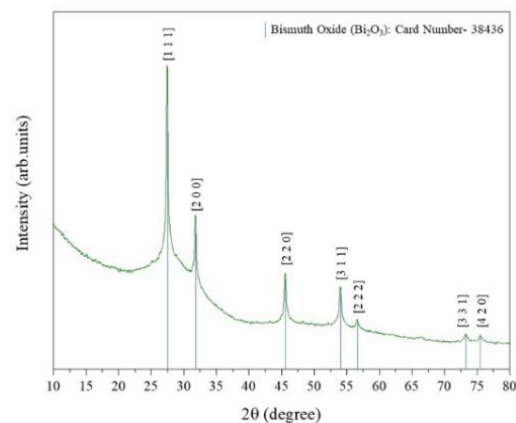


Figure 2. The X-ray diffraction spectra for the glass ceramic sample

Table 1. The density (D) and molar volume (V_m) of the glass ceramic $30\text{Li}_2\text{O} : 3\text{MoO}_3 : 40\text{Bi}_2\text{O}_3 : 20\text{TeO}_2 : 7\text{CuO}$

$D \text{ (g/cm}^3\text{)}$	$V_m \text{ (cm}^3\text{/mol)}$
4.53	52.39

Table 2. The electrical resistivity (ρ), the Seebeck coefficient (S), and the power factor (PF) with measurement temperature (T) of the sample

$T \text{ (K)}$	$\rho \text{ (}\Omega\text{m)}$	$S \text{ (V/K)}$	$PF \text{ (W/mK}^2\text{)}$
315	3.83	0.04	3.46×10^{-4}
377	3.82	-0.001	3.57×10^{-7}
425	0.00	0.01	Inf.
474	1.24	-0.09	6.99×10^{-3}
523	3.42	0.02	1.66×10^{-4}
571	1.87	0.02	1.87×10^{-4}

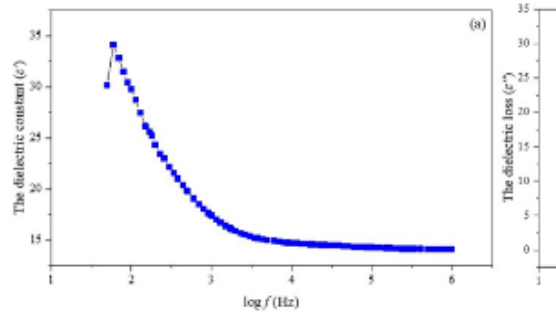


Figure 2. The AC measurements for the glass ceramic sample; (a) the dielectric constant

The conclusion reached indicates that electrons (e^-) are the charge carriers. The mechanism of electrical conduction in amorphous materials is based on the Davis-Mott model. The conduction occurs by hopping when the excited carrier from the valence band's edge to the gap's localized tail states at moderate temperatures. Electrons are examples of localized charge carriers that tend to influence their surroundings. Many physical atoms can experience these kinds of aberrations. The carrier's energy is often decreased by distortion. The result is that the carrier is trapped inside itself due to the attraction exerted (Marshall and Owen, 1971; El-Mallawany, 1995). This conclusion corresponds to the research results of the electrical resistivity (ρ) value, shown in Table 2 as the reciprocal of the electrical conductivity value. Hence, the electrical characteristics of glass ceramic materials exhibit values solely within a particular temperature range rather than tending to grow or decrease with the temperature used to determine them. Calculating the thermoelectric efficiency in this work shows that the highest power factor follows Equation (1).

$$PF = S^2 \rho^{-1} \quad (1)$$

Where S is the Seebeck coefficient, and ρ is the electrical resistivity. It was shown on the highest value of the power factor 6.99 mW/mK^2 at a temperature of 473 K. From Table 2, it can be seen that the electrical resistivity (ρ) at 425 K is $0 \text{ } \Omega\text{m}$ and the Seebeck coefficient (S) is 0.01 V/K . According to Equation (1), if ρ value is $0 \text{ } \Omega\text{m}$, PF will become infinity. The value of electrical resistivity is $0 \text{ } \Omega\text{m}$ at the investigated temperature of 425 K is attributed to the unique structural characteristics and conduction mechanism of the glass ceramic material, which obtain a phase transition upon changes in the temperature, pressure, or other outside factors (Gonon and Dupla, 2021). In this experiment, the change in testing temperature induced a phase transition within the

material's internal structure, leading to a significant decrease in electrical resistivity. The researchers have referenced this phenomenon from relevant research studies in response to the reviewer's question and will use these questions as a basis for further development of oxide glass ceramic thermoelectric materials in future work.

Conclusions

Glass ceramic samples in the composition of 30Li₂O: 3MoO₃: 40Bi₂O₃: 20TeO₂: 7CuO were prepared by the traditional melt quench technique. The Bi₂O₃ phase was investigated by X-ray diffraction. Dielectric loss (ϵ'') is reduced within the measuring frequency range according to the defined dielectric characteristics. The electrical properties were investigated on the electrical resistivity, the Seebeck coefficient, and the power factor (PF) with increasing temperature measurement. The highest efficiency of thermoelectric property in this work is shown on the highest power factor $6.99 \times 10^{-3} \text{ (W/mK}^2\text{)}$ at 474 K. Since this glass ceramic has a high degree of stability and efficiency, it may be used in thermoelectric devices for cooling systems and power production.

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