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A study in thermoelectric effect of Eu_2O_3 substitution on Al_2O_3 in $CuAl_{(100-x)}Eu_{(x)}O_2$

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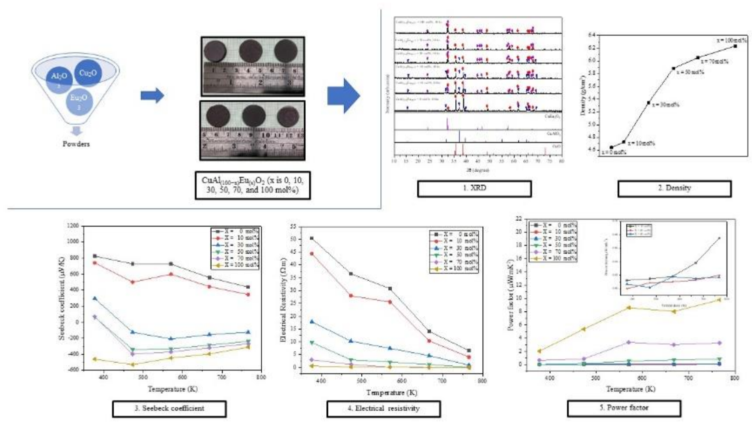
Abstract

The effects of Eu_2O_3 substitution on Al_2O_3 in $CuAl_{(100-x)}Eu_{(x)}O_2$ (x = 0, 10, 30, 50, 70, and 100 mol%) were considered. In this study, all samples with a diameter of 20 mm were sintered by using solid-state reaction method. The structural analysis with XRD investigation shows that it had a hexagonal $CuAlO_2$ structure at x = 0, 10, 30, and 50 mol%. Tetragonal $CuEu_2O_4$ structures were reported at x = 10, 30, 50, 70, and 100 mol%. The ZEM-3 instrument measures thermoelectric characteristics like the Seebeck coefficient, electrical resistivity, and power factor throughout the temperature range of 350 − 800 K. According to the investigation, the electrical resistivity value tended to continue to decrease as measurement temperature increased. The Seebeck coefficient shows a positive value. However, when Al_2O_3 was substituted for Eu_2O_3 in large amounts, the Seebeck coefficient value became negative. The power factor increases as the mole ratio of Eu_2O_3 increase, exhibited the highest value of around 9.50 µW mK^{-2} at 775 K.



Keywords: Solid-state reaction; Seebeck coefficient; Power factor

Introduction

Because waste heat can be converted to electrical energy, the development of thermoelectric materials is critical for convert energy. Improved thermoelectric material performance is a crucial factor to take into account. The performance of thermoelectric materials can be identified using a parameter called the figure of merit, $Z = \sigma\alpha^2/\kappa$, where σ, α, and κ are the electrical conductivity, Seebeck coefficient, and thermal conductivity, respectively [1]. Large values of Z are indicators of good thermoelectric material performance, and it is important to get high σ, high α, and low κ values.

For industries that work in high temperature situations some thermoelectric materials, such as SnSe [2, 3], PbTe [4, 5], and Bi-Te [6, 7] have performance limitations as a result of this. At a temperature of 300 K, I. Terasaki et al. reported a $NaCo_2O_4$ thermoelectric material with a high Z value of 8.8×10^{-4} and a big S of 100 µV K^{-1} [8]. The usage of $NaCo_2O_4$ is nonetheless constrained by sodium's inability to withstand temperatures higher than 1073 K and by air humidity [9]. As a result, high-performance and environmentally stable new oxide materials must be created. Creating novel oxide materials remains the most critical challenge for practical thermoelectric power-generating applications [10-11]. $CuAlO_2$ compounds, particularly illustrate excellent performance for thermoelectric materials that operate at high temperatures. At high temperatures, Park et al. [12] and Liu et al. [13] found that the temperature independence of $CuAlO_2$ thermoelectric characteristics for Seebeck coefficient, thermal conductivity, power factor, and ZT is about 5 $\mu V\ K^{-1}$, 0.20 $W\ cmK^{-1}$, 4×10^{-5} $W\ mK^{-2}$, and 0.0045, respectively. Chattopadhyay et al. [14] were investigated the thermoelectric and electrical characteristics of $CuAlO_2$ thin films produced with dc-sputtering. It is generally established that altering the microstructure and carrier concentration by substitution is a suitable technique of improving thermoelectric performance.

The thermoelectric characteristics of Eu_2O_3 substitution on Al_2O_3 in $CuAl_{(100-x)}Eu_{(x)}O_2$ (x = 0, 10, 30, 50, 70, and 100 mol%) pellet of ceramics with a diameter of 20 mm synthesized by the solid-state reaction process are investigated. The thermoelectric properties of the samples: electrical resistivity, Seebeck coefficient, and power factor were investigated by ZEM-3. The results of this study are likely to assist in the optimization of thermoelectric materials $CuAlO_2$.

Materials and Methods

When x is 0, 10, 30, 50, 70, and 100 mol%, we should calculate using the stoichiometry method in the chemical formula $CuAl_{(100sx)}Eu_{(x)}O_2$. Cu_2O with a purity of 99.99%, Al_2O_3 with a purity of 99.99%%, and Eu_2O_3 with a purity of 99.99% were utilized in this method for the calculated post-material synthesis. This work synthesized the material in an electric furnace by a solid-state reaction method. The powder of the chemical mixture was calcined at 373 K for 2 hr to remove organic matter and moisture. The calcined powder was ball milled for 3 hr at 200 rpm with agate balls as the grinding medium. The dry powder was pressed using a hand press at a pressure of 300 MPa to prepare 20 mm diameter pellets. All pellets were sintered inside a furnace at 1173 K for 10 hr in air, and then the furnace was cooled down to room temperature.

X-ray diffraction was used to determine the samples' phases (XRD; Shimazdu XRD-6100) with CuKá radiation accelerated in a 2θ range of 10 − 80 degrees. The Archimedes principle of density (A&D HR-200) was used to confirm the physical characteristics. The Seebeck coefficient, electrical resistivity, and power factor were measured (ZEM-3; Ulvac, Inc.) in the temperature rang 350 − 800 K.

Results and Discussions

All of X-ray diffraction patterns of $CuAl_{(100-x)}Eu_{(x)}O_2$ (x is 0, 10, 30, 50, 70, and 100 mol%) samples follow the molar ratio of Eu_2O_3 from 0 until 1 by mol% were shown in Fig. 1. These results presented the primary phase in CuO (Monoclinic, JCPDS No.00-048-1548) for all samples. $CuAlO_2$ (Hexagonal, JCPDS No.00-040-1037) for samples with increased Eu_2O_3 of 0.1, 0.3, and 0.5 mol % and $CuEu_2O_4$ (Tetragonal, JCPDS No.00-052-1719) for all samples except those without Eu_2O_3. The amplitude of the $CuEu_2O_4$ peak was observed to increase as the Eu_2O_3 concentration increased. Throughout the 350 K to 800 K temperature range, the ZEM-3 was utilized to measure thermoelectric properties.

In Fig. 2, the density is shown on the y-axis as the Eu_2O_3 content increases on the x-axis. The sample density measurement indicates the decreasing impact of Al_2O_3 and the increasing amount of Eu_2O_3 in the $CuAlO_2$ and $CuEu_2O_4$ phases, and Eu_2O_3 has a higher density than Al_2O_3. The increase in Eu_2O_3 content inside the sample causes an increase in the density value. The material with the most excellent density was 6.24 g cm^{-3} of Eu_2O_3 in mol%.

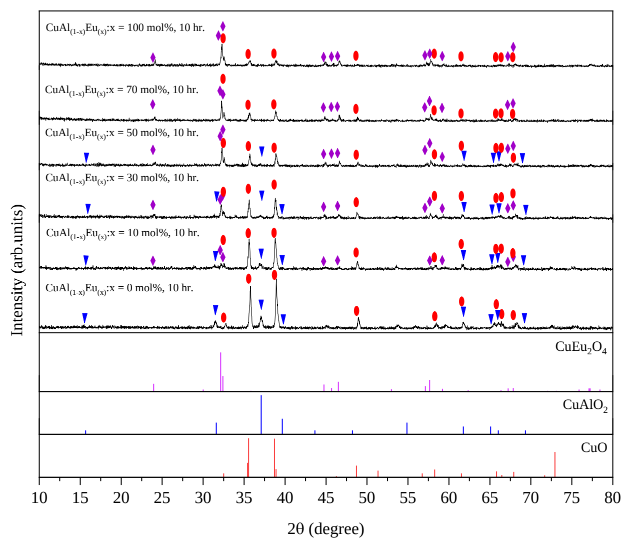


Fig. 1 X-ray diffraction patterns of $CuAl_{(100-x)}Eu_{(x)}O_2$ (x is 0, 10, 30, 50, 70, and 100 mol%).

The Seebeck coefficient's sign, which is positive for the conductivity of holes and negative for the conductivity of electrons, reflects the electrical conductivity of the material under investigation. The Seebeck coefficients for all samples with increased Eu_2O_3 from x = 0 to x = 100 mol% are shown in Fig. 3. The conductivity behavior of materials was changed from hole conductivity to electron conductivity [15]. The electrical conductivity of a system increases as the

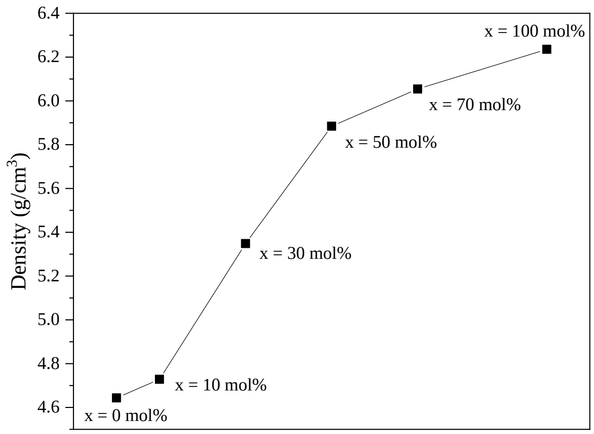


Fig. 2 Density of $CuAl_{(100-x)}Eu_{(x)}O_2$ (x is 0, 10, 30, 50, 70, and 100 mol%).

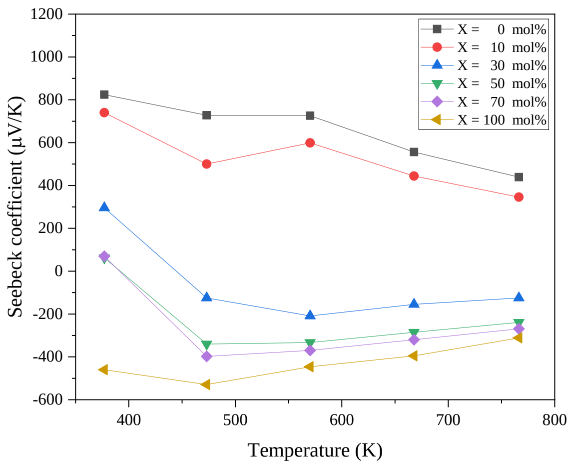


Fig. 3 Temperature dependence on Seebeck coefficient of $CuAl_{(100-x)}Eu_{(x)}O_2$ (x is 0, 10, 30, 50, 70, and 100 mol%).

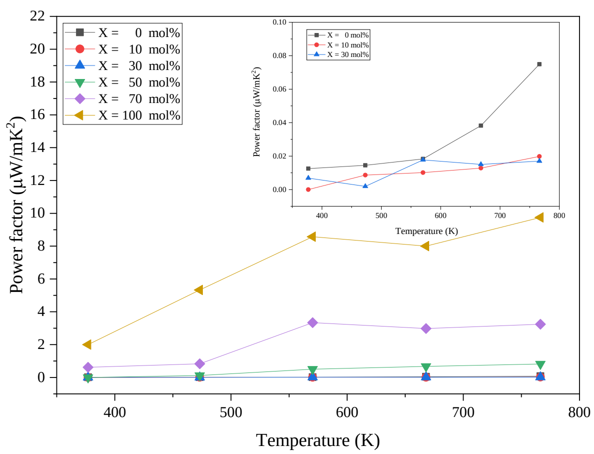


Fig. 5 Temperature dependence on Power factor of $CuAl_{(100-x)}Eu_{(x)}O_2$ (x is 0, 10, 30, 50, 70, and 100 mol%).

carrier density raises. The Seebeck coefficient in the measured sample was determined to be trending downward when the measurement temperature was raised. The sample where x is 0 mol% at 377 K has the highest value of 824 μV K^{-1} for the measured Seebeck coefficient. The Mott relation can be used to explain the Seebeck coefficients tendency [16].

$$S(T) = \frac{\pi^2 k_B^2 T}{3e}\left[\frac{N(E)}{n} + \left(\frac{\partial \ln \mu(E)}{\partial E}\right)_{E=E_F}\right] \quad (1)$$

where are Seebeck coefficient (S), the Boltzmann constant (k_B), carrier concentration (n), and density of state ($N(E)$), respectively. Fig. 4 shows that when the molar ratio of Eu_2O_3 rises, the electrical resistivity of $CuAl_{(100-x)}Eu_{(x)}O_2$ samples decreases with increasing temperature throughout the whole temperature range. The electrical resistivity ρ and the Seebeck coefficient S were used to analyses the power factor $P = S^2/\rho$. Fig. 5 shows the power factor calculated from the data in Figure 3 and 4. With increasing temperature, the power factor of the $CuAl_{(100-x)}Eu_{(x)}O_2$ samples tends to rise. The highest value of the sample is defined as 9.75 μW mK^{-2} when x is 1 at 775 K after determining the power factor that tends to rise with increasing temperature utilized in the measurement as shown in Fig. 5. Based on the findings, it's thought that polycrystalline $CuAl_{(100-x)}Eu_{(x)}O_2$ might be useful in high temperature thermoelectric applications.

Conclusion

The main phase present in the sintered bulk samples of $CuAl_{(100-x)}Eu_{(x)}O_2$ ($0 \leq x \leq 1$) is Tetragonal structure, I4/mmm, combination with other phases such as CuO and $CuAlO_2$. With the ZEM-3 instrument, the measuring temperature range of 350 − 800 K was determined to assess the thermoelectric characteristics. The $CuAl_{(100-x)}Eu_{(x)}O_2$ samples Seebeck coefficient reduced as the Eu_2O_3 concentration increased, result of an increase in carrier density. The electrical resistivity decreases with increasing temperature throughout the whole temperature range. Because of a consider the effects in electrical conductivity, adding Eu_2O_3 up to x = 100 mol% resulted in a significant rise in power factor. The highest value of power factor 9.75 μW mK^{-2} on x is 100 mol% of Eu_2O_3 at 775 K. As a result, adding Eu_2O_3 to $CuAlO_2$ improved the thermoelectric properties of the material at high temperatures.

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