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Research Article

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Effect of Manufacturing Process to Physical Properties of Thermoelectric $CuAlO_2$ Ceramics

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Abstract This paper presents the improvement of conventional technology in fabricating of $CuAlO_2$ thermoelectric materials. The phase $CuAlO_2$ is formed at 1423K. At higher temperatures, a part of phase $CuAlO_2$ is separated into Al_2O_3 and CuO. In addition, final materials with improved technology for a single phase, high conductivity and Seebeck coefficient is more stable than materials created with the traditional process.

Keywords $CuAlO_2$, thermoelectric, creating process

Introduction

Thermoelectric materials can convert thermal energy directly to electrical energy via the Seebeck effect. The performance of the thermoelectricmaterialcan be evaluated by the figure of merit, $Z = S^2\sigma / \kappa$, where S, σ and κ are the Seebeck coefficient, electrical conductivity and thermal conductivity, respectively [1]. High thermoelectric efficiency requires high power ($PF = S^2\sigma$) and low thermal conductivity. However, it is difficult to control these parameters independently becauseSeebeck coefficient is inversely proportional to the electrical conductivity. Besides, when the thermal conductivity was reduced, the electrical conductivity decreased as well. Therefore, the study of thermoelectric materials must aim to increase the electric conductivity and reduce the thermal conductivity. High temperature operation also raised conversion efficiency of thermoelectric system. Further attention has been put on oxide ceramics as thermoelectric materials because they are very suitable for long term use at high temperature in air.

For years, thermoelectric effect have attracted many scientific considerations of both basic researches and applications. The research aims is looking for materials to converting heat energy into electricenergy with the highest efficiency. Some oxide systems have been investigated such as $Ca_3Co_4O_9$ [2], ZnO [3], $SrTiO_3$ [4],... These materials have many advantages, such as high thermal and chemical stability, low cost, and easy creation. Recently, another metal oxide thermoelectric material, $CuAlO_2$, also received concentration due to its high temperature stability, high thermoelectric efficiency [5]. Ishiguro et al have extensively studied the crystalline structure of $CuAlO_2$. Koumotoet al firstreported $CuAlO_2$ with a power factor ~ 1.04×10^{-4} $Wm^{-1}K^{-2}$for single crystal and ~ 2.0×10^{-5} $Wm^{-1}K^{-2}$ at 1073K forpolycrystalline $CuAlO_2$, respectively [6]. Park and co – workers reported power factors at 1140K for $CuAlO_2$ ceramics sintered at 1433 K and 1473K, respectively 4.98×10^{-5} and 6.62×10^{-5} $Wm^{-1}K^{-2}$[5]. Therefore, $CuAlO_2$ceramics is a promising material for thermoelectric energy conversion applications.

In 2014, $CuAlO_2$ thermoelectric materials were researched in Hue and achieved some initial results [7]. However, the properties and stability are low as the manufacturing technology has not been fully studied. This paper presents the improvement of traditional ceramic technology in synthesizing $CuAlO_2$thermoelectric materials at Solid State Physics department, College of Science, Hue University.

Materials and Methods

All samples $CuAlO_2$ were prepared from the reagent – grade powders: CuO (95%, Korea) and Al_2O_3 (98%, Korea). The mixture powders were milled in ethanol for 20 hours. After the powders had been dried, pressed to prepare pelletsand calcined at 1073K for 2 hours to create in $CuAlO_2$ phase. The formed mixture was mill again for 20 hours and pressed into pellets with 100MPapressure and sintered at 1423K for 2 hours. The specimens for electrical measurements were in the form of rectangular bars of 2 mm × 2 mm × 15 mm. Sintered samples were further annealed at temperature in range (573 ÷ 973) K for 4 hours before analyzing proccess. The crystalline phase was analyzed using an X – ray diffractometer (XRD, D8 ADVANCE). Thermoelectric properties were analyzed using a thermoelectric system of Nguyen TrongTinh et al from Institute of Applied Physics and Scientific Instrument of Vietnamese Academy of Science and Technology [8].

Results and Discussion

Phase $CuAlO_2$was synthesized from CuO and Al_2O_3 by solid state reaction method following reaction chain [5]:

$CuO + Al_2O_3 \rightarrow CuAl_2O_4$

$CuAl_2O_4 + CuO \rightarrow 2CuAlO_2 + 1/2O_2$

The XRD profiles of sintered $CuAlO_2$were annealing atseveral temperatures 573K, 673K, 773K, 873K, 973Kfor 4 hours are shown in Figure 1.

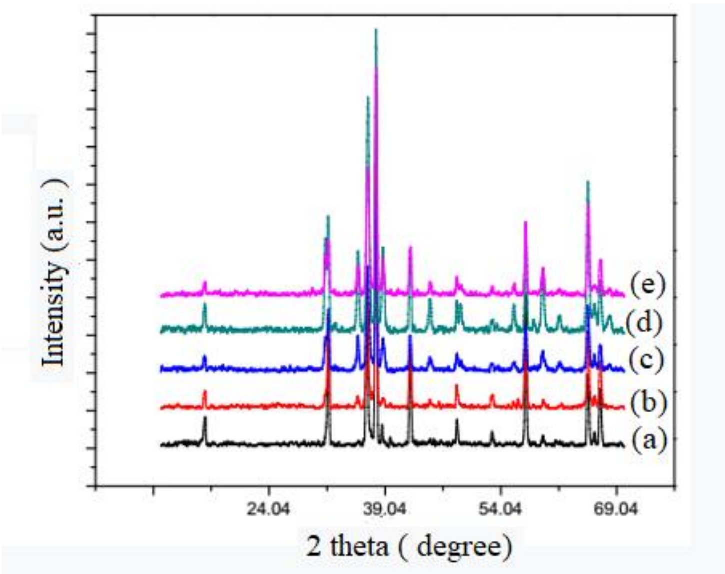

Figure 1: The XRD pattern of the $CuAlO_2$sample annealed at the different temperature: (a)573K, (b) 673K, (c) 773K,(d) 873K, (e) 973K.

Figure 1 shows X-ray diffraction (XRD) patterns at room temperature with of $CuAlO_2$ samples annealed at different temperatures of 573 K, 673 K, 773 K, 873 K, and 973 K in 4 hours. It can be seen that annealed $CuAlO_2$ samples at temperatures of 573 K, 673 K, 773 K, and 873 K the phase of $CuAlO_2$ was created almost completely, only a very small amount of $CuAl_2O_4$ exists. However, for the incubation sample at 973 K besides $CuAlO_2$ phase, there is mainly the appearance of Al_2O_3, CuO and Cu_2O phase. The ratio of the second phase increased when the annealing temperature was above 873 K. In our opinion, there has been partial separation of $CuAlO_2$ phase to produce Al_2O_3 and CuO at high annealed temperature. Therefore, the temperature in range from 573 K to 873 K was be chosen for further annealing process.

The material density are listed in Table 1.The temperature dependence of density for $CuAlO_2$ annealed from 573K and 873K is shown in Figure 2.

Table 1: The influence of $CuAlO_2$ density (ρ) of on annealing temperature

T (K)	ρ (g/cm^3)
573	4.66
673	4.69
773	4.59
873	4.61

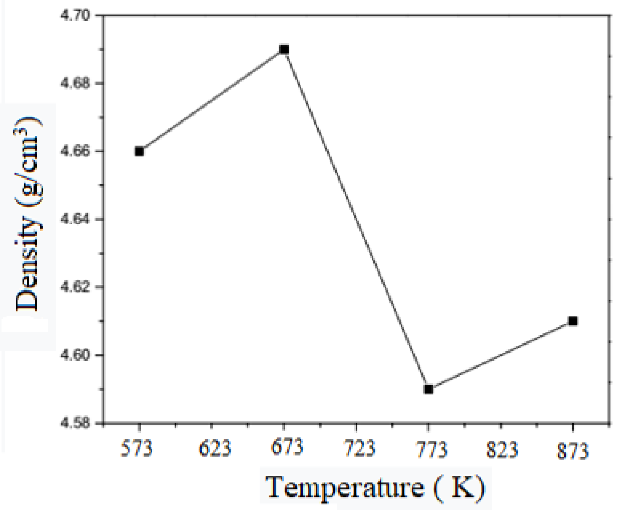

Figure 2: The annealed temperature dependence of $CuAlO_2$ densities

Table 1 shows that the ceramic density increases when the annealing temperature rises and reaches a highest value at 673K. Then, the density decreases with further increasing annealing temperature. According to the structural analysis, with annealing temperatures above 873K, a part of $CuAlO_2$ material is split into CuO and Al_2O_3.The percentage of these phases increases more with higher annealing temperature, causing the homogeneity of chemical composition in materials. This may cause a decrease in the proportion of ceramics when the annealing temperature is increased after sintering.

To investigate the temperature dependence of thermoelectric properties, we survey about the variation of thermoelectric parameters at different annealing temperature.

Figure 3 shows the variation of resistivity for $CuAlO_2$ceramics with several annealing temperatures at773K, 873K, and 973K.

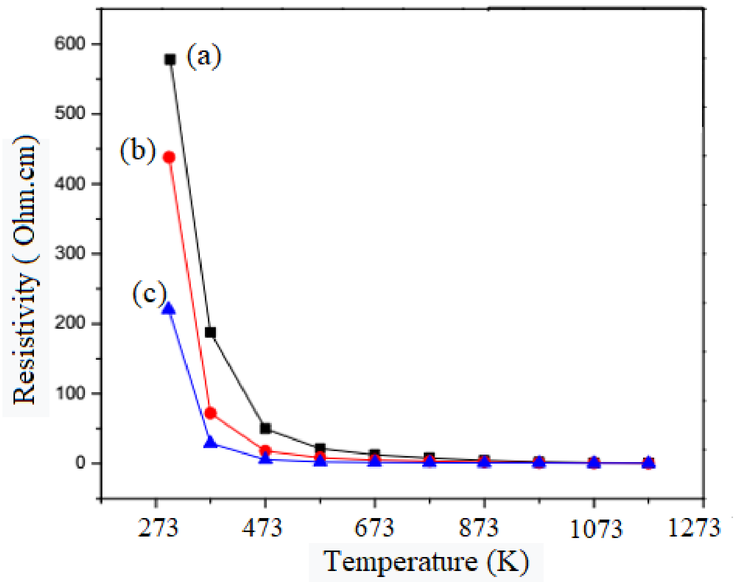

Figure 3: The temperature dependence of resistivity in $CuAlO_2$ annealed at (a) 773K, (b) 873K, (c) 973K

Figure 3 shows that the resistivity of the sample decreases with increasing annealing temperature. Thehigherannealingtemperature,the lower resistivity is. Thus, the annealing temperature affects thermoelectric properties of sintered $CuAlO_2$ ceramics.

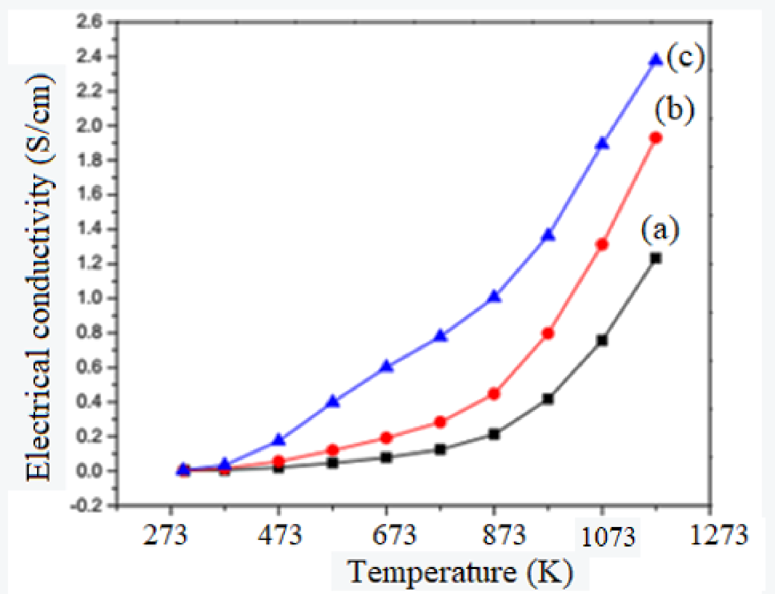

Figure 4: The temperature dependence of electricalconductivity in $CuAlO_2$ annealed at (a) 773K, (b) 873K, (c) 973K.

Figure 4 indicated when the annealing temperature increase, the electrical conductivity of the material increases. However, the speed and value of the electrical conductivity of the sample annealed at 973K is much larger than that at 773K and 873K. It is possibly b3 in range of annealed temperatures above 873K, a part of $CuAlO_2$ material is split into CuO and Al_2O_3. Thus, it can be assert that annealing temperatures affect the electrical conductivity of sintered $CuAlO_2$ thermoelectric ceramics.

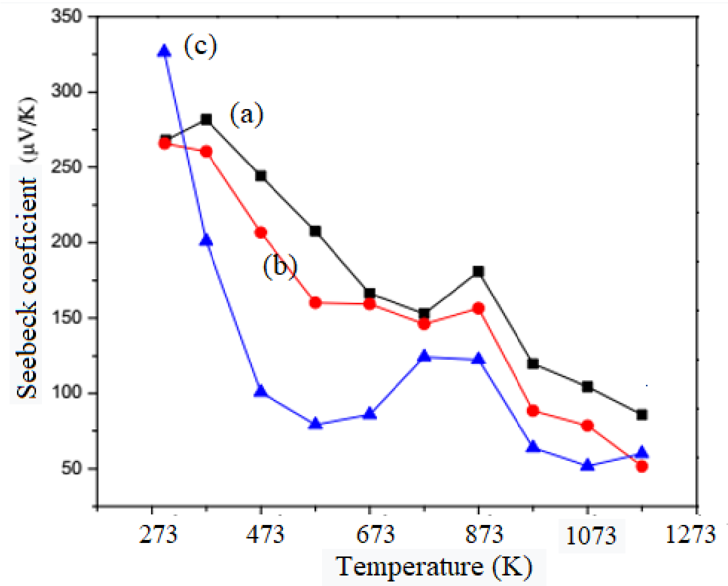

Figure 5: Seebeck coefficient of $CuAlO_2$ samples as a funtion of annealing temperature at (a) 773K, (b) 873K, (c) 973K

The variation of Seebeck coefficients with temperature of $CuAlO_2$ ceramics annealed at 773K, 873K, and 973K. Accordingly, the values of the Seebeck coefficient in $CuAlO_2$samplesannealed at higher temperature tend to decrease. Especially,at 773Kannealing temperature, the variation of Seebeck coefficient with temperature reaches the maximum value. On the contrary, phase separation is a significant impact on the thermoelectricproperties of the ceramicin high temperature range.

Conclusion

By solid state reaction method and improvement process, we have synthesized the $CuAlO_2$ ceramics at differentfurther annealed temperatures. The annealing temperature were chosen in range from 673K to 873K. At this temperature range, it is possible to obtain a single phase ceramics, with higher electrical conductivity and more stable Seebeck coefficient than that made by conventional method.

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