Electrical Characteristics $CuFe_2O_4$ Ceramics With and Without Al_2O_3 for Negative Thermal Coefficient (NTC) Thermistor

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ABSTRACT

In order to get capability in thermistor production in Indonesia, a study on electrical characterization of $CuFe_2O_4$ base-ceramics with Al_2O_3 addition has been performed. The Al_2O_3 addition was done with various concentrations namely 0, 1 dan 5 mole %. Powder of CuO, Fe_2O_3 and Al_2O_3 was mixed and ground. The mixture was pressed with pressure of 3,9 ton/cm^2 to form pellets. The pellets was then sintered at 1100^{0}C for 2 jam in air. After sintering, two sides of some sintered pellets were coated with silver paste and fired at 600^{o}C for 10 minutes. Some of coated samples were heat treated at 500^{o}C for 5 minutes in N_2 gas. These pellets were analyzed using x-ray diffraction (XRD). R-T and ageing characteristics were evaluated. From the XRD data, it was known that the $CuFe_2O_4$ ceramics produced crystallized in tetragonal spinel. According to the electrical data, the thermistor constant (B) and sensitivity (a) increases due to the addition of Al_2O_3. It was known from the ageing test that only the ceramics with 0 and 1 mole % Al_2O_3 fit the electrical stability condition.

Keywords : *Ceramic, $CuFe_2O_4$, Al_2O_3, thermistor, NTC, ageing.*

1. INTRODUCTION

NTC thermistor are widely used due to its capability to be applied in many applications such as temperature sensor, electric current limiter, flowrate meter and pressure sensor[1]. It is known that generaly the NTC thermistor is made of ceramic having structure of spinel of AB_2O_4 where A is the ion occupies tetrahedarl position and B is the ion occupies octrahedral position[2-10]. Many works have been performed in order to improve the characteristic of the NTC thermistor having spinel structure [6,7,11]. Theoritically, the addition of additive such as Al_2O_3 may change the electrical characteristics of $CuFe_2O_4$ ceramic.

When the additive of Al_2O_3 is added to the $CuFe_2O_4$ ceramic, the characteristics of the $CuFe_2O_4$ ceramic may cange because two conditions may accur namely, the first, Al_2O_3 dissolves in the $CuFe_2O_4$ through substituting Cu ions or Fe ions, the second, Al_2O_3 does not dissolve and segregate at grain boundaries. When the substitution of Fe or Cu ions results in an increase of electron in the conduction band, the electrical resistivity of the $CuFe_2O_4$ will decrease. On the contrary, when the second condition occurs, the electrical resistivity may increase due to a change in microstructure. In this work, the effect of the Al_2O_3 addition on the electrical characteristics, especially the electrical stability, of the $CuFe_2O_4$ ceramic was studied.

2 . METHODOLOGY

Powder of CuO, Fe_2O_3 and aditive of Al_2O_3 (0, 1 and 5 mole %) were mixed and ground. The composition is shown at Table 1. After calcination at 800^{o}C for 2 hours, the mixture was ground. The ground powder was pressed with pressure of 3,9 ton/cm^2 to form pellets. The green pellets were sintered at 1100^{0}C for 2 hours in furnace air. Two sides of some sintered pellets were coated with silver paste. Some silver coated samples were heat treated at 500^{o}C for 5 minutes in N_2 gas. Structure of the sintered pellet was analyzed using x-ray diffraction (XRD). For electrical characterization, the electrical resistance of the pellets was measured at various temperatures. The measurement was done before and after ageing test. The ageing test was conducted by measuring the resistance of the pellet at room temperature after heating at 150^{o}C in every several hours.

Thermistor constant (B) is the gradient of the ln Resistivity vs 1/T curve which is constructed based on equation (1) [2-11] expressing NTC characteristic. Sensitivity (α) was calculated using equation (2)[6].

$$R = R_0.\ \text{Eksp.}(\frac{B}{T}) \ldots\ldots..(1)$$

where R = thermistor resistance (Ohm), R_0 = a constant (Ohm), B = Thermistor constant (K) and T = Temperature (K).

$$\alpha = \frac{-B}{T^2} \ldots\ldots\ldots\ldots(2)$$

where α = Sensitifitas termistor,

B = Koefisien termistor dalam ^oK

T = suhu dalam ^oK

Activation energy can be calculated using equation (3)[6],

$$B = \frac{\Delta E}{k} \ldots\ldots..(2)$$

Dengan B = Konstanta termistor (^oK)

ΔE = Energi aktivasi (eV),

k = Konstanta Boltzmann ($\frac{eV}{^oK}$)

Table 1. Sample composition in mole %.

No.	CuO	Fe_2O_3	Al_2O_3
1.	40	60	0
2.	40	59	1
3.	40	55	5

4. RESULTS AND DISCUSSION

4.1 XRD Analyses

XRD profiles of $CuFe_2O_4$ base-ceramics added with Al_2O_3 are shown at Fig. 1 and Fig.2 as the representative. All the $CuFe_2O_4$ base-ceramics crystallized in tetragonal structure (JCPDS No.34-0425). In all XRD profiles peak of Fe_2O_3 was observed indicating that a part of the Fe_2O_3 could not form $CuFe_2O_4$ solid solution. Peak from Al_2O_3 was not observed in all XRD profiles. This fact gives a possibility that the Al_2O_3 was dissolved.

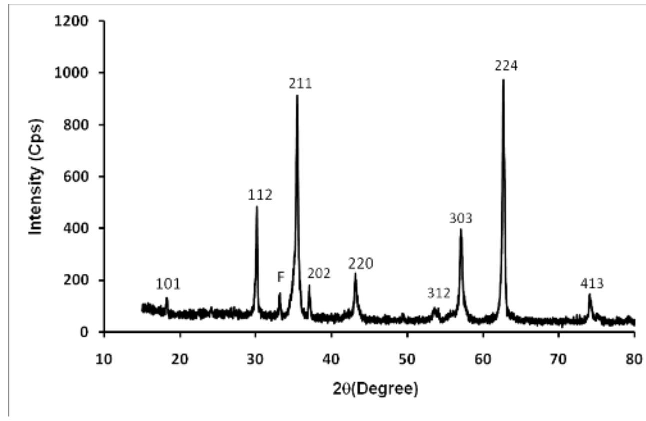

Fig.1. XRD profile of $CuFe_2O_4$ base-ceramic (40CuO-60Fe_2O_3) without Al_2O_3 addition. F is peak from Fe_2O_3.

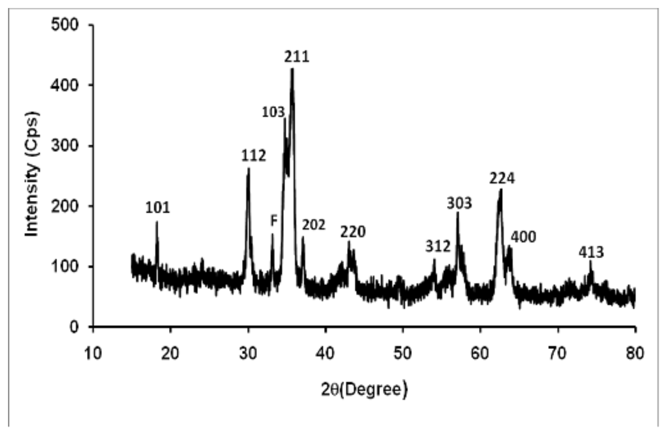

Fig.2. XRD profile of $CuFe_2O_4$ base-ceramic (40CuO-60Fe_2O_3) with 1 mole % Al_2O_3. F is peak from Fe_2O_3.

4.2 Electrical Characteristics

Curves of ln Resistivity vs 1/T in Fig. 3 are linear indicating that the $CuFe_2O_4$ base-ceramics obey the NTC characteristic of the thermistor. As shown in Table 1, thermistor constant (B) and sensitivity (alpha) of the ceramics added with Al_2O_3 are larger than those of the ceramics without Al_2O_3. This means that the addition of Al_2O_3 icreased the thermistor constant and sensitivity of the ceramics. Large B and α is good for the NTC thermistor. The value of B and α of the ceramics fit the market requirement (larger than or equal 2000K(B) and 2.2%/K(α,alpha)). The mechanism of the increasing the thermistor constant and sensitivity is as follow. The Al_2O_3 segregates at grain boundaries and impeded grain growth during sintering. The ceramics then contains small grains or many grain boundaries. Because the grain boundaries are scattering center for electron, the electrical resistivity and thermistor constant of the ceramics increase.

The ageing test result is shown in Fig. 4. As can be seen, for sample without Al_2O_3, from 0 to 100 hours ageing test, the resistivity change rapidly following the aging time. From 100 hours to 1000 hours, the resistance tends to stable. Below 100 hours, the ions in the ceramics tend to rearrange during heating at 150°C[12]. Here, some Fe^{2+} oxidizes to Fe^{3+}. So, the resistance has not been stable. In the range 100-1000hours, the resistance tends to stable though slightly fluctuates. The time to reach a stability condition is different depending on the concentration of Al_2O_3. For sample added with Al_2O_3 the time to reach the stable condition is about 200 hours. It is clear that the addition Al_2O_3 worsen the stability. The fluctuation of the resistance becomes larger as the concentration of Al_2O_3 increases. The value of 200 hours can be taken as a time for preparing a stable $CuFe_2O_4$ base-ceramics for NTC thermistor.

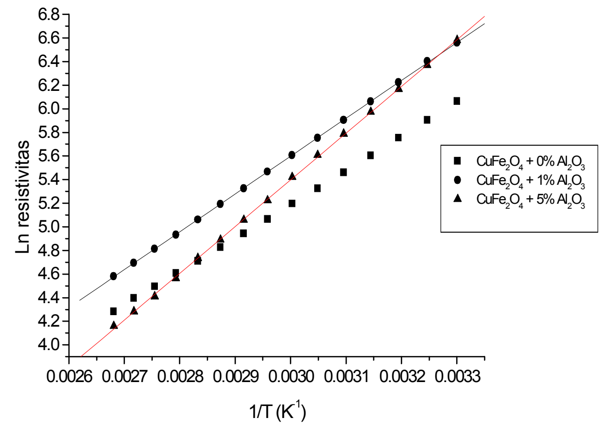

Fig.3. The relation between ln Electrical Resistivity and 1/T.

Table 1. Electrical characteristics of the ceramics.

No.	Penambahan Al_2O_3 (mole %)	B (^{0}K)	α (%/ ^{0}K)
1.	0	2862	3,22
2.	1	3208	3,61
3.	5	3958	4,46

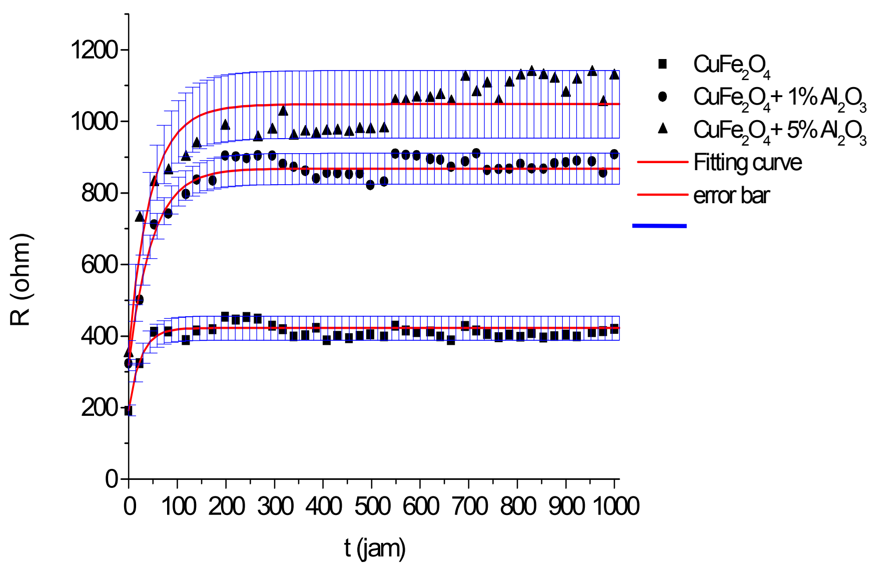

Fig. 4. Electrical resistance as function of time as the result of ageing test.

5. CONCLUSION

All $CuFe_2O_4$ base-ceramic crystallized in tetragonal structure. Thermistor constant (B) and sensitivity (a) of the $CuFe_2O_4$ base-ceramics increase with the increase of Al_2O_3 concentration. This means that the additionof Al_2O_3 can be used as a controlling parameter. However, the addition of Al_2O_3 decreases the electrical stability of the $CuFe_2O_4$ base-ceramics. Only sample without Al_2O_3 and that added with 1 mole % Al_2O_3 fit the electrical stability condition. Heating at 150°C for 200 hours can be used to make $CuFe_2O_4$ base-ceramic stable electrically.

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