Electrical Characteristics of $CuFe_2O_4$ Thick Film Ceramics With Different Glass Frit Concentrations Fired at 1000 ^{o}C for Negative Thermal Coefficient (NTC) Thermistor

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ABSTRACT

Fabrication of $CuFe_2O_4$ thick film ceramics utilizing Fe_2O_3 derived from yarosite using screen printing technique for NTC thermistor has been carried out. Effects of glass frit concentration (0; 2,5 ;5 weight %) have been studied. X-ray diffraction analyses (XRD) was done to know crystal structure and phases formation. SEM analyses was carried out to know microstructure of the films. Electrical properties characterization was done through measurement of electrical resistance at various temperature (room temperature to 100^{o}C). The XRD data showed that the films crystalize in tetragonal spinel. The SEM images showed that the effects of glass frit concentration make the grain size was smaller. Electrical data showed that the larger the glass frit concentration, the larger the : resistance, thermistor constant and sensitivity. From the electrical characteristics data, it was known that the electrical characteristics of the $CuFe_2O_4$ thick film ceramics followed the NTC characteristic. The value of B and R_{RT} of the produced $CuFe_2O_4$ ceramics namely B = 2215-2807^{o}K and ρ_{RT} = 6,9-16,7 M Ohm, fitted market requirement.

Key Words

Ceramics, $CuFe_2O_4$, Thick Film,Thermistor, NTC, Glass Frit.

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 glass frit concentrations may change the electrical characteristics of $CuFe_2O_4$ ceramic.

NTC thermistors are widely used in the world and the need for them is still increasing due to its potential use for many sectors such as biomedical, aerospace, instrumentation, communications, automotive and HVACR (Heating, Ventilation, Air conditioning and Refrigeration) [Betatherm, 2008, Na, et al., 2001].

Some areas for NTC thermistor applications are temperature measurement, circuit compensation, suppression of in rush-current, flow rate sensor and pressure sensor [Park and Han, 2005]. It is traditionally known that most NTC thermistors are produced from spinel ceramics based on transition metal oxides forming general formula AB_2O_4 where A is metal ion in tetrahedral position and B is metal ion in octahedral position [Na, et al., 2001, Park, 2003, Matsuo, 1982, Jung, 1993, Hamada, 2001, Park and Han, 2005, Park and bang, 2003, Fritsch, 2004, Schmidt, 2004]. One of the spinel ceramics that can be used for NTC thermistor is $CuFe_2O_4$.

In Indonesia many electronic components are generally imported. In order to decrease the dependency of Indonesia to the imported product and get capability in self producing the NTC thermistor as well as increase the added value of mineral abundant in Indonesia, a study on fabrication of $CuFe_2O_4$ NTC thermistor in the form of pellet or disk has been carried out by utilizing yarosite as raw material [Wiendartun, 2007, Wiendartun, 2008]. However, the application of the $CuFe_2O_4$ thermistor in the form of pellet is more limited. In order to extend the area of application, it is required to also produce the thermistor in another form that more practical and profitable economically. Here, the form of thick film is considered as the choice. The thermistor in the form of thick film is possible for miniaturization and integration.

Technology generally used for fabrication of thick film thermistor is screen printing which is technically simple. Some parameters in the screen printing namely viscosity of paste, screen size, paste composition and firing parameter such as time and temperature significantly influence the characteristic of the thick film produced. In this work, a study on fabrication of thick film thermistor based on $CuFe_2O_4$ with different glass frit concentration was performed. The effect of different glass frit concentration on the characteristics, especially the electrical characteristics, of the $CuFe_2O_4$ thick film ceramics for NTC thermistor is the focus of the study.

2 . METHODOLOGY

Powder of Fe_2O_3 derived from yarosite mineral (chemical composition is shown in Table 1) and glass frit made of SiO_2, B_2O_3 and PbO were crushed and sieved with a sieve of 38 μm (Hole size of 38μm). The method for processing the Fe_2O_3 powder is described elsewhere [Wiendartun,2007]). The sieved Fe_2O_3 powder and (0; 2,5 ;5 weight %) glass frit was mixed. The mixture of Fe_2O_3 and glass frit was mixed with organic vehicle containing alpha terpineol and ethyl cellulose with composition of 90 weight % and 10 weight %, respectively, to form a paste. The paste was screen printed on alumina substrates using screen printing technique. The films were fired at 1000°C for 1 hour in air. The crystal structure of the fired thick films was analyzed with x-ray diffraction (XRD) using Kα radiation. The films were investigated by SEM to evaluate their microstructure (morphology). A couple of parallel electrodes which is 1 mm apart are made on the sensor side of the fired thick film by using Ag paste. After the paste was dried at room temperature, the Ag coated-thick films were heated at 600°C for 10 minutes. The resistance was measured at various temperatures from 25 to 100°C in steps of 5°C using a digital multimeter and a laboratory made chamber equipped with a digital temperature controller. Thermistor constant (B) was derived from Ln resistivity vs. 1/T curve where B is the gradient of the curve based on (1)[Park, 2003,Park and Han, 2005]:

$$\rho = \rho_o .\exp (B/T) \qquad (1)$$

where, ρ is the electrical resistivity, ρ_o is a constant or the resistivity at T is infinite, B is the thermistor constant and T is the temperature in Kelvin.

Room temperature resistance (R_{RT}) was determined as the electrical resistance at room temperature (25°C) and sensitivity (α) was calculated using (2) [Moulson and Herbert, 1990].

$$\alpha = B/T^2 \qquad (2)$$

where, α is the sensitivity, B is the thermistor constant and T is the temperature in Kelvin.

Microstructure and structural analyses were carried out by using a Scanning Eectron Microscopy (SEM) and X-ray diffraction (XRD), respectively. All steps of the procedure can be seen in Fig. 1.

Table 1. Chemical composition of Fe_2O_3 powder derived from yarosite.

Component	Concentration (Weight %)
Fe_2O_3	93.80
SiO_2	1.15
Al_2O_3	2.54
TiO_2	1.02
MgO	0.19
MnO	0.09
K_2O	0.12
Na_2O	0.50
CaO	0.59

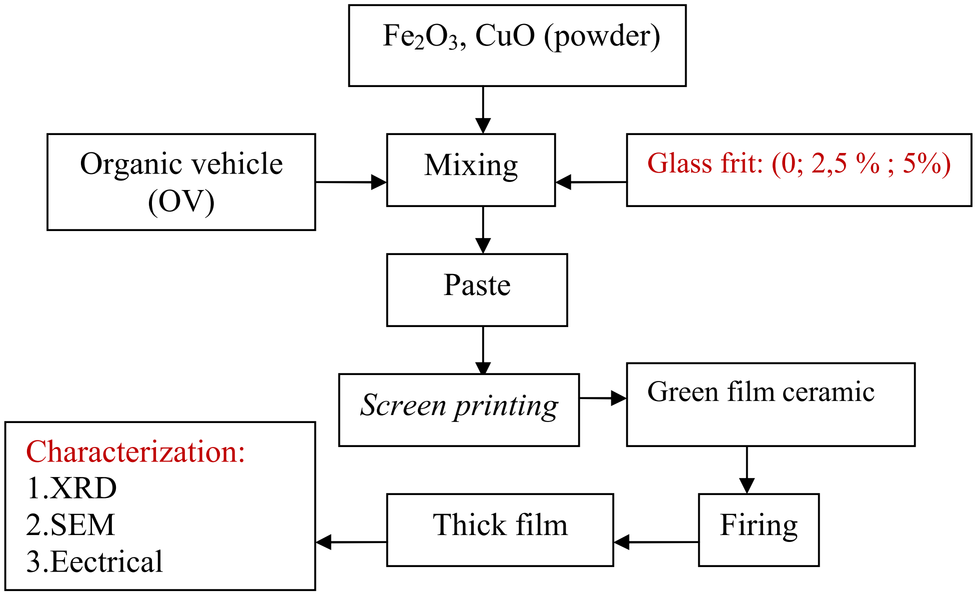

Fig.1. Flow diagram of the experiment procedure.

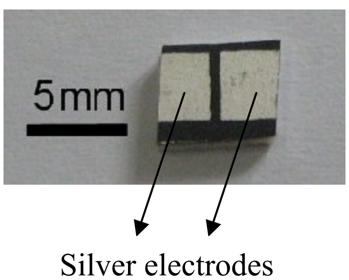

Fig.2. A typical thick film thermistor.

3. RESULTS AND DISCUSSION

Fig1 shows the experiment principle for electrical characterization. Fig. 2 shows the appearance a typical thick film. Fig.3, Fig.4 and Fig.5 shows the XRD profiles of $CuFe_2O_4$ thick film ceramics fired at 1000^oC for 1 hour and different glass frit concentration at 0; 2,5 and 5 % respectively. As shown in the figure Fig.3, Fig.4 and Fig.5 the profiles are similar. The XRD profiles show that the structure of the thick film ceramics is tetragonal spinel after being compared to the XRD standard profile of $CuFe_2O_4$ from JCPDS No. 34-0425). No peaks from second phases observed. It may be due to the small concentration of impurities which is smaller than the precision limit of the x-ray diffractometer used.

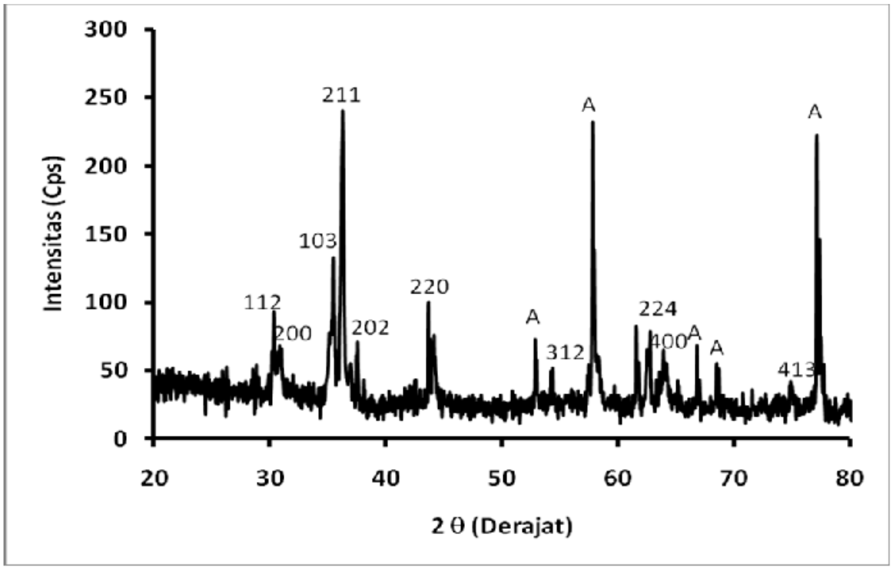

Fig.3. XRD profile of $CuFe_2O_4$ based-thick film fired at 1000^{o}C for 1 hr
(without glass frit concentration)

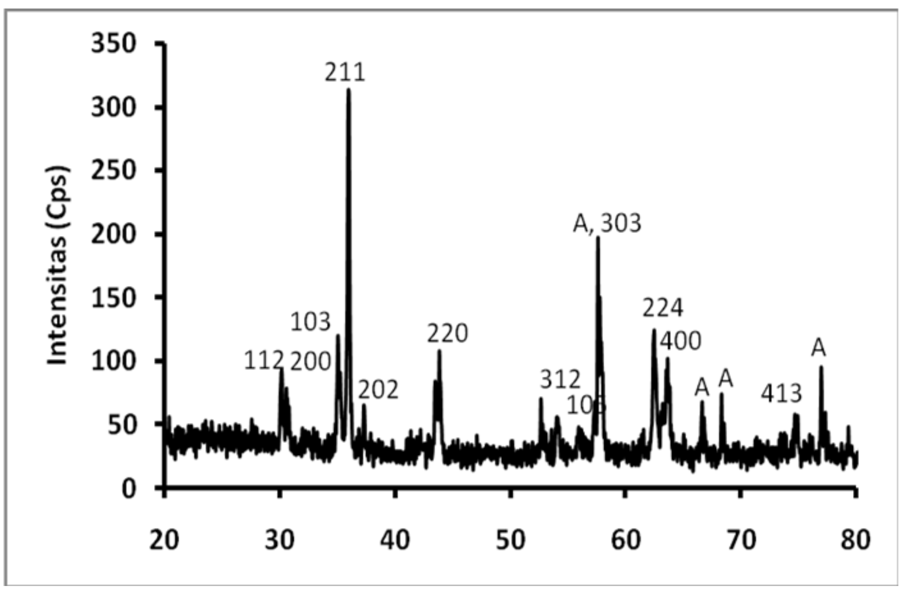

Fig.4 XRD profile of $CuFe_2O_4$ based-thick film fired at 1000^{o}C for 1 hr
(with 2,5 % glass frit concentration)

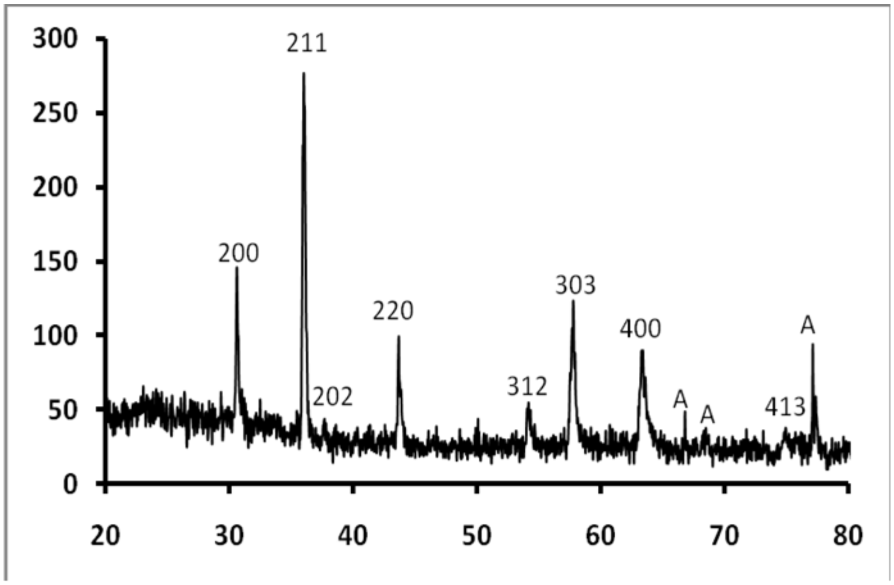

Fig.5. XRD profile of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hr (with 5,0 % glass frit concentration)

Microstructures of the $CuFe_2O_4$ film ceramic fired at 1000°C for 1 hour with : 0; 2,5 , 5% glass frit concentration respectively, are depicted in Fig.6, and Fig. 7. All of the thick films are characterized in porous structure with different grain size depending on the glass frit concentration. The grain size becomes smaller following the increasing of the glass frit concentration. This is a consequence of the smaller mobility of ions at the increase glass frit concentration. According firing glass frit, delayed grain growth.

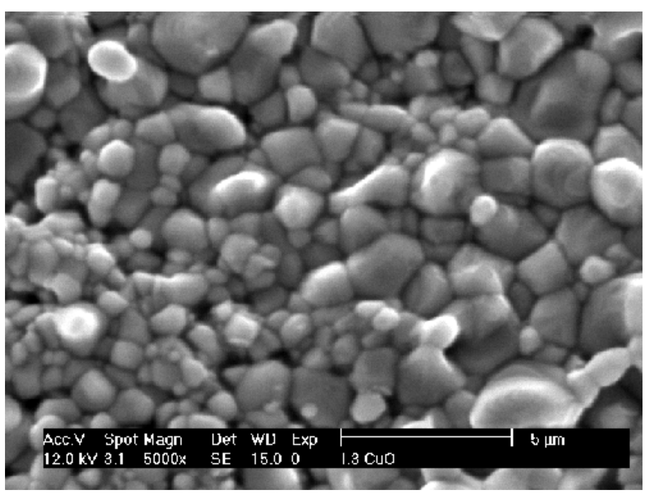

Fig.6 Microstructure of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hr (without glass frit concentration)

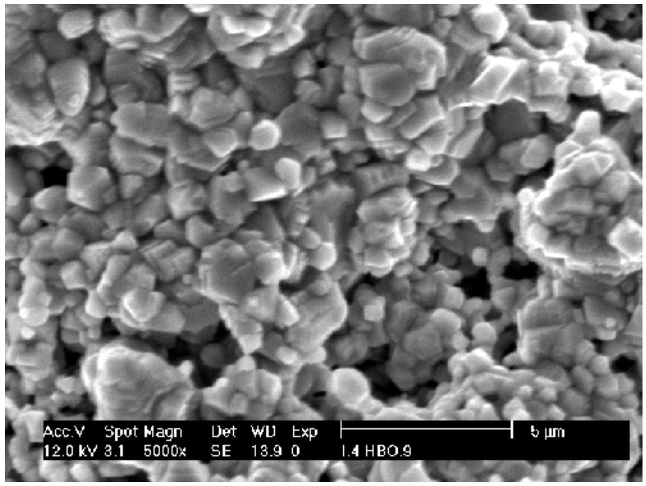

Fig.7 Microstructure of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hr
(with 2,5 % glass frit concentration)

The electrical data of the $CuFe_2O_4$ thick film ceramics is shown in Fig. 8 and Table 2. The electrical data of Fig. 8 shows that the Ln resistivity increases linearly as the $1/T$ increasing, indicating that the electrical characteristics of the ceramics follows the NTC tendency expressed by equation (1). As shown in Table 2, The increase of the glass frit concentration from 0% to 5% increases the room temperature resistance (R_{RT}), thermistor constant (B) and sensitivity (α). The value of thermistor constant (B) ceramics fitted market requirement. (B >= 2000K).

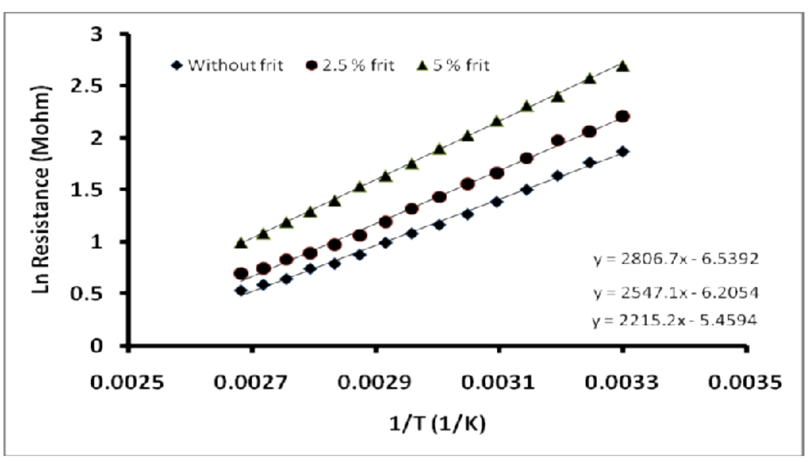

Fig.8. The relation between ln Electrical Resistivity and 1/T of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hr
(with : 0; 2,5 , 5% glass frit concentration)

Table 2. Electrical characteristics of the $CuFe_2O_4$ based-thick film fired at 1000^oC for 1 hr (with : 0; 2,5 , 5% glass frit concentration)

No.	Frit (%)	B (K)	Alfa (%/K)	R_{SR} (Mohm)
1.	0	2215	2,46	6,9
2.	2,5	2547	2,83	9,8
3.	5	2807	3,12	16,7

CONCLUSION

$CuFe_2O_4$ thick film ceramics utilizing Fe_2O_3 derived from yarosite mineral have been well fired at 1000^oC for 1 hour. with : 0; 2,5 , 5% glass frit concentration. All of the thick films crystallize in tetragonal spinel. The SEM images showed that the effects of glass frit concentration make the grain size was smaller. Electrical data showed that the larger the glass frit concentration, the larger the : resistance, thermistor constant and sensitivity. From the electrical characteristics data, it was known that the electrical characteristics of the $CuFe_2O_4$ thick film ceramics followed the NTC characteristic. The value of thermistor constant (B) = 2215-2807^oK and room temperature resistance (R_{RT}) = 6,9-16,7 MOhm of the produced $CuFe_2O_4$ ceramics fitted market requirement.

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