Journal of Materials Science Research; Vol. 1, No. 3; 2012
ISSN 1927-0585 E-ISSN 1927-0593
Published by Canadian Center of Science and Education

Electrical Characteristics $Cufe_2o_4$ Thick Film Ceramics with Different Glass Concentrations Fired at 1000 °C for Negative Thermal Coefficient (Ntc) Thermistor

Wiendartun[1] & Dani Gustaman Syarif[2]

[1]Department of Physics Education, Education University of Indonesia (UPI), Bandung, Indonesia

[2]PTNBR-BATAN, Jl.Tamansari 71, Bandung, Indonesia

Correspondence: Department of Physics Education, Education University of Indonesia (UPI), Jl. Dr. Setiabudhi 229, Bandung, Indonesia. E-mail: wien_upi@yahoo.com

Received: March 5, 2012 Accepted: March 23, 2012 Online Published: June 10, 2012

doi:10.5539/jmsr.v1n3p70 URL: http://dx.doi.org/10.5539/jmsr.v1n3p70

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. Effect of glass frit addition (0, 2.5, 5 weight %) has been studied. X-ray diffraction analyses (XRD) was done to know crystal structure and phases formation. SEM analyses were carried out to know microstructure of the films. Electrical properties characterization was done through measurement of electrical resistance at various temperatures (room temperature to 100°C). The XRD data showed that the films crystalize in tetragonal spinel. The SEM images showed that glass frit addition made the grain size 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^oK and ρ_{RT} = 6,9-16,7 M Ohm, fitted market requirement.

Keywords: ceramics, $CuFe_2O_4$, thick film, thermistor, NTC, glass frit

1. Introduction

NTC thermistor is widely applied in many applications such as temperature sensor, electric current limiter, flowrate meter and pressure sensor (Anonymous, 2008). It is generaly known that the NTC thermistor is made of ceramic having structure of spinel of AB_2O_4 where A is the ion occupies tetrahedral position and B is the ion occupies octrahedral position (Fritsch, 2004; Hamada, 2001; Jung, 1993; Matsuo, 1982; Moulson & Herbert, 1990; Na et al., 2001; Park, 2003; Park & Bang, 2003; Park & Han, 2005; Schmidt et al., 2004; Wiendartun & Syarif, 2007; Wiendartun et al., 2008; Wiendartun & Syarif, 2008). Many efforts can be performed in order to improve the characteristic of the NTC thermistor having spinel structure. One of the spinel ceramics may be applied as NTC thermistor is $CuFe_2O_4$ ceramic. The thermistor may be produced in the form of disk or thick film. Here, the object of study is the thick film thermistor.

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. Experimental Procedures

Powder of Fe_2O_3 derived from yarosite mineral (chemical composition is shown in Table 1), CuO powder 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 & Syarif, 2007). The sieved Fe_2O_3 and CuO powder and (0, 2.5, 5 weight %) glass frit were mixed. The mixture of Fe_2O_3, CuO 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. 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 & Han, 2005):

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

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.

Table 1. Chemical composition of Fe_2O_3 powder derived from yarosite

Component	Concentration (Weight %)
Fe2O3	93.80
SiO2	1.15
Al2O3	2.54
TiO2	1.02
MgO	0.19
MnO	0.09
K2O	0.12
Na2O	0.50
CaO	0.59

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

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

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 Electron Microscopy (SEM) and X-ray diffraction (XRD), respectively. All steps of the procedure can be seen in Figure 1.

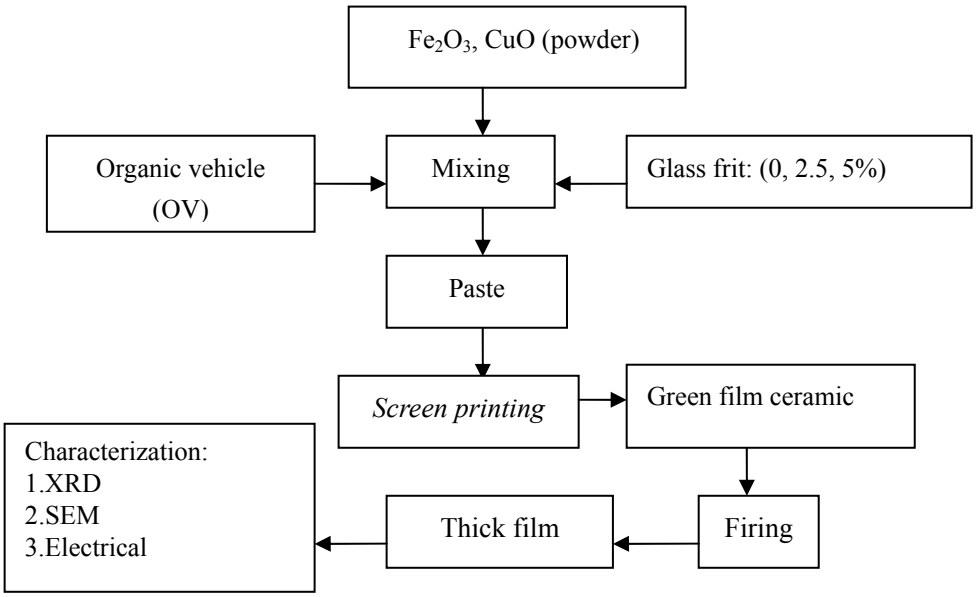

Figure 1. Flow diagram of the experiment procedure

3. Results and Discussions

Figure 2 shows the appearance a typical thick film. Figure 3, Figure 4 and Figure 5 show the XRD profiles of $CuFe_2O_4$ thick film ceramics fired at 1000°C for 1 hour with different glass frit concentrations of 0; 2,5 and 5 %, respectively. As shown in the figure Figure 3, Figure 4 and Figure 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. Peaks from alumina substrate (with A sign) are observed.

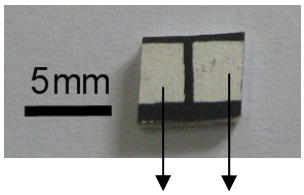

Figure 2. A typical thick film thermistor

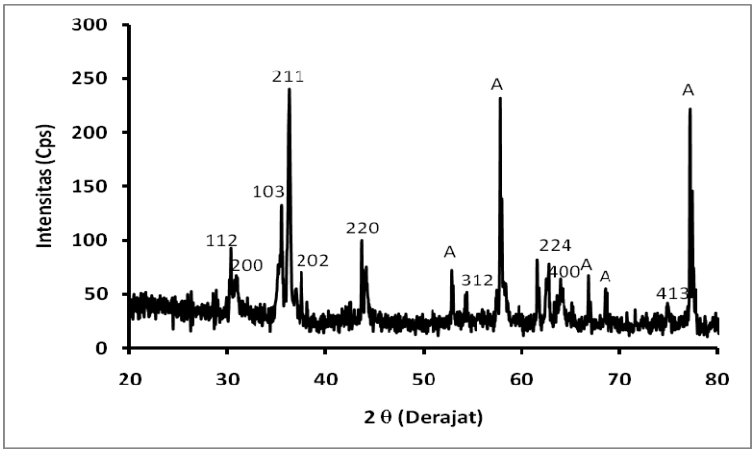

Figure 3. XRD profile of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hour (without glass frit addition)

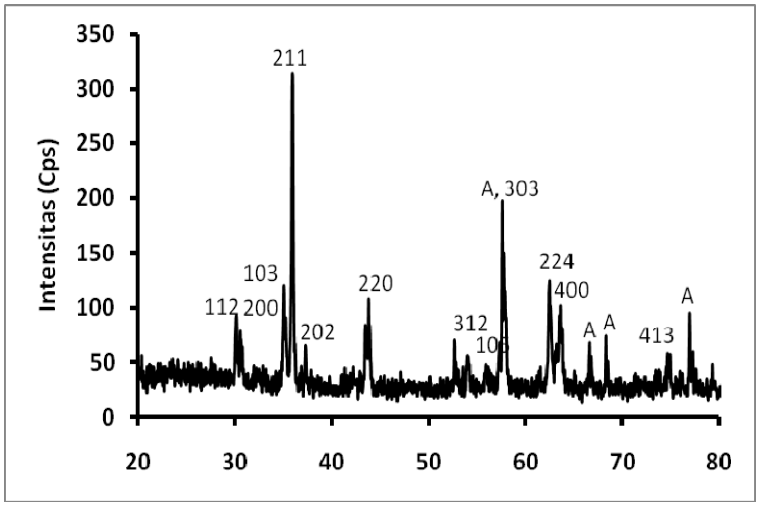

Figure 4. XRD profile of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hour (with 2.5 % glass frit addition)

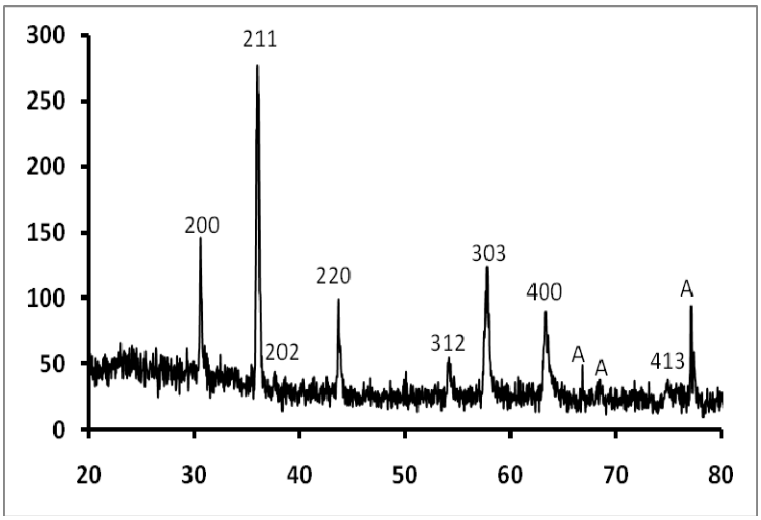

Figure 5. XRD profile of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hour (with 5.0 % glass frit addition)

Microstructures of the $CuFe_2O_4$ film ceramic fired at 1000°C for 1 hour with 0 and 2.5 % glass frit concentration respectively, are depicted in Figure 6, and Figure 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 due to the increase of glass frit concentration.

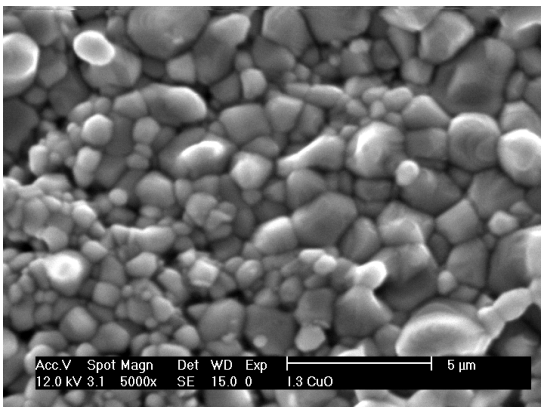

Figure 6. Microstructure of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hour (without glass frit addition)

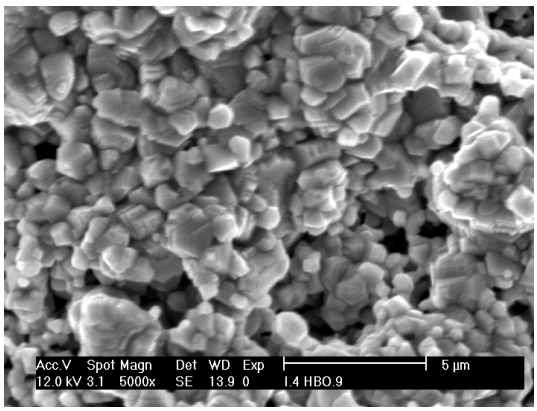

Figure 7. Microstructure of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hour (with 2.5 % glass frit addition)

The electrical data of the $CuFe_2O_4$ thick film ceramics is shown in Figure 8 and Table 2. The electrical data of Figure 8 shows that the Ln resistivity increases linearly as the $1/T$ increases, 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 thermistor constant (B) for market requirement is ≥ 2000K. So, the value of thermistor constant (B) of the ceramics in this work fitted market requirement.

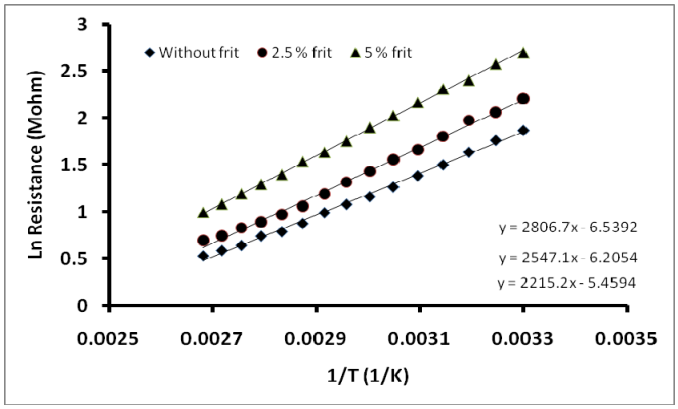

Figure 8. The relation between ln Electrical Resistivity and 1/T of $CuFe_2O_4$ based-thick film fired at 1000°C for 1 hour with 0, 2.5 and 5% glass frit addition

Table 2. Electrical characteristics of the $CuFe_2O_4$ based-thick films fired at 1000°C for 1 hour with: 0, 2.5 and 5% glass frit concentration

No.	Frit(%)	B(K)	α (%/K)	R RT (Mohm)
1.	0	2215	2.46	6.9
2.	2.5	2547	2.83	9.8
3.	5	2807	3.12	16.7

4. Conclusion

$CuFe_2O_4$ thick film ceramics utilizing Fe_2O_3 derived from yarosite mineral have been well fired at 1000°C for 1 hour with 0, 2.5 and 5% glass frit addition. All of the thick films crystallize in tetragonal spinel. Glass frit addition makes grain size of the films smaller. The larger the glass frit concentration, the larger the resistance, thermistor constant and sensitivity. The electrical characteristics of the $CuFe_2O_4$ thick film ceramics follow the NTC characteristic. The value of thermistor constant (B) = 2215-2807^oK and room temperature resistance (R_{RT}) = 6,9-16,7 MOhm. Compared to the value of B for market requirement namely ≥ 2000K, the B value of the produced $CuFe_2O_4$ ceramics fits market requirement.

Aknowledgments

The authors wish to acknowledge their deep gratitude to Directorate General of Higher Education(DIKTI), Ministry of National Education of Indonesian Government for financial support under *HIBAH BERSAING* program with contract No. 2784/H.40/PL/ 2009, 07 Mei 2009

References

Anonymous. (2008). Retrieved from http://www.betatherm.com

Fritsch S. G., Sarrias, S. J., Rousset, A., Schuurman, S., & Lannoo, A. (2004). Mechanical properties of nickel manganites-based ceramics used as negative temperature coefficient thermistors. *Materials Research Bulletin, 39*, 1957-1965. http://dx.doi.org/10.1016/j.materresbull.2004.05.020

Hamada, K., & Oda, H. (2001). Thermistor composition. *US Patent*, 6,270,693, August 7.

Jung, H. J., Yoon, S. O., Hong, K. Y., & Lee, J. K. (1993). Metal oxide group thermistor material. *US Patent*, 5,246,628, September 21.

Matsuo, Y., Hata, T., & Kuroda, T. (1982). Oxide thermistor composition. *US Patent*, 4,324,702, April 13.

Moulson, A. J., & Herbert, J. M. (1990). *Electroceramics*. New York: Chapman & Hall, p.140.

Na, E. S., Paik, U. G., & Choi, S. C. (2001). The effect of a sintered microstructure on the electrical properties of a Mn-Co-Ni-O thermistor. *Journal of Ceramic Processing Research, 2*(1), 31-34.

Park, K. (2003). Microstructure and electrical properties of $Ni_{1.0}Mn_{2-x}Zr_xO_4$ ($0 \leq x \leq 1.0$) negative temperature coefficient thermistors. *Materials Science and Engineering, B104*, 9-14. http://dx.doi.org/10.1016/S0921-5107(03)00257-5

Park, K., & Bang, D. Y. (2003). Electrical properties of Ni-Mn-Co-(Fe) oxide thick film NTC thermistors, *Journal of Materials Science: Materials in Electronics, 14*, 81-87. http://dx.doi.org/10.1023/A:1021900618988

Park, K., & Han, I. H. (2005). Effect of Al_2O_3 addition on the microstructure and electrical properties of $(Mn_{0,37}Ni_{0,3}Co_{0,33}\text{-}_xAl_x)O_4$ ($0 \leq x \leq 0.03$) NTC thermistors. *Materials Science and Engineering, B119*, 55-60. http://dx.doi.org/10.1016/j.mseb.2005.01.018

Schmidt, R., Basu, A., & Brinkman, A. W. (2004). Production of NTCR thermistor devices based on $NiMn_2O_{4+\delta}$, *Journal of The European Ceramic Society, 24*, 1233-1236. http://dx.doi.org/10.1016/S0955-2219(03)00415-1

Wiendartun, & Syarif, D. G. (2007). Characterization of $CuFe_2O_4$ ceramics added with Al_2O_3 for NTC thermistor using Fe_2O_3 derived from yarosite (In Bahasa Indonesia), p.80. *In Proceedings of the National Seminar of Nuclear Science and Technology*, Bandung, Indonesia.

Wiendartun, & Syarif, D. G. (2008). Synthesis and Characterization of $CuFe_2O_4$ Thick Film Ceramics for NTC Thermistor Using Yarosite Mineral as Raw Material. *Proceeding on The International Seminar on Chemistry (ISC-2008) UNPAD Bandung*, Indonesia, October, 30-31.

Wiendartun, Syarif, D. G., Suhendi, E., Sambodo, G. D., & Setiawan, A. (2008). Synthesis and characterization of $CuFe_2O_4$ ceramics for NTC thermistor by utilizing Fe_2O_3 derived from yarosite mineral (In Bahasa Indonesia), p.110. *In Proceedings of The National Seminar of Ceramics*, Bandung, Indonesia.