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The Effect of Various Sintering Condition to the Electrical Characteristics of Fe$_2$TiO$_5$ Ceramics for NTC Thermistor

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Abstract. Iron Titanium Oxide (Fe$_2$TiO$_5$) ceramics for negative temperature coefficient (NTC) thermistor have been synthesized in various sintering condition in order to know the effect of sintering condition, namely, temperature, time and atmosphere condition, on the characteristic especially the electrical characteristic of Fe$_2$TiO$_5$ ceramics. Temperature conditions are adjusted between 1200 and 1300$^\circ$C, for 2 and 8 hours in furnace with atmospheric air and oxygen (O$_2$) gas. For Fe$_2$TiO$_5$ ceramics sintered in the air, the effect of sintering temperature on the grain size was clearly seen. With increasing sintering temperature and sintering time, grain size of Fe$_2$TiO$_5$ ceramics increased. However, the effect of atmospheric O$_2$ gas to their grain size is not clearly seen although it has reduced tendency. XRD analysis showed that all ceramics Fe$_2$TiO$_5$ are single phase of orthorhombic structure. From electrical resistances data that was measured at temperature 40 – 250$^\circ$C, it is found that the value of thermistor constant (B), activation energy (E$_a$) and room temperature resistance (R$_{RT}$) decreases with increasing sintering temperature and sintering time. But the value of B, E$_a$, and R$_{RT}$ increases with atmospheric O$_2$ gas condition.

1. Introduction

NTC thermistor has been widely used around the world today due to its capability in various fields of electronics, such as thermometer, electric current limiter, water flow sensor, and pressure sensor [1-2]. Fe$_2$TiO$_5$ is one of semiconductor ceramics used as based material for main components fabrication of NTC thermistor as temperature sensor. The composition of mineral Fe$_2$TiO$_5$ belongs to pseudobrokyte group where the general formula of this compound is X$_2$YO$_5$ with octahedral in both side, X and Y. Generally, Fe$_2$TiO$_4$ mostly used for gas sensor, nonlinear optic, magnetic, catalyst, and microelectronics [3-4]. Since Fe$_2$TiO$_4$ actually has semi-conductivity, it is very capable to use Fe$_2$TiO$_4$ for NTC thermistor as based material. Thermistor constant $B$ is a quantity which determine typical characteristic of thermistor corresponding to electrical resistance changes with temperature. The larger thermistor constant leads the better thermistor quality. Many works and studies have been conducted by researchers to enhance thermistor constant $B$ and thermistor sensitivity $\alpha$. They have studied the effect of sintering time to typical characteristic of Fe$_2$TiO$_5$-based ceramics. The research of NTC thermistor for high temperature has been reported [5]. However, the reports of NTC thermistor in high temperature fabricated by Fe$_2$TiO$_5$-based ceramics have not been excessively reported so far. Recently, electrical characterizations of Fe$_2$TiO$_5$-based ceramics for NTC thermistor with concentration 0, 0.4, and 1.0 % mole of each MnO$_2$ dopant sintered at 1100$^\circ$C for 90 minutes in air...
have been carried out [6]. Through that research, it is found that thermistor constant (B) of Fe$_2$TiO$_5$-based ceramics increased along with increasing of MnO$_2$ dopant concentration. The obtained value of constant thermistor is 5018–6476 K. It significantly shows that thermistor can be used in accordance with market need which is $\geq 2000$ K [7-10]. Beside dopant concentration, in order to obtain optimum condition, sintering parameter (temperature, time, and atmosphere) should be varied since they will give effect to electrical characteristic of obtained ceramics. Therefore, the aim of this research is to investigate the effect of sintering parameter on their electrical characteristic of Fe$_2$TiO$_5$-based ceramics for NTC thermistor.

2. Materials and Method

Fe$_2$TiO$_5$ thermistor ceramic was prepared by using commercial powder of Fe$_3$O$_4$ and TiO$_2$. Mixture of Fe$_2$O$_3$ and TiO$_2$ with composition of 50:50 in mole %, respectively, to form pellets, calcined at 700$^\circ$C for 2 hours. In order to form pellets, pressed powder, the homogeneous mixture of Fe$_2$O$_3$ and TiO$_2$ was pressed at 4.10$^7$ kg/m$^2$. The raw pellets were sintered at 1100-1300$^\circ$C for 2-8 hours in air and oxygen (O$_2$) atmosphere. Pictures of sintered pellets were taken to know their visual appearance. The crystal structure of the sintered pellets analyzed with x-ray diffraction (XRD) using K$_\alpha$ radiation at 40 kV in voltage and 25 mA in current. The pellets ceramic were investigated by SEM to evaluate their microstructure (morphology). The resistance was measured at various temperatures from 40 to 250$^\circ$C in steps of 10$^\circ$C using a digital multimeter and a laboratory made chamber equipped with a digital temperature controller. Both surfaces of sintered pellets had been coated by conductive silver paste colloid silver solution and heated at 600$^\circ$C for 10 minutes in advance. Thermistor constant (B) was derived from Ln resistivity vs. $1/T$ curve where B is the gradient of the curve based on equation 1 [11-13]:

$$ R = R_0 \exp (B/T) $$

Where, $R$ is the electrical resistance, $R_0$ is a constant or the resistant at the infinite temperature, B is the thermistor constant and $T$ is the temperature in Kelvin and k is the Boltzmann constant. Room temperature resistance ($R_{RT}$) was determined as the electrical resistance at room temperature (25$^\circ$C).

From the value of B, the activation energy ($E_a$) and sensitivity ($\alpha$) were calculated using equation 2 and 3 [14-16].

$$ E_a = B.k $$

$$ \alpha = B/T^2 $$

3. Results and Discussion

The XRD profiles of Fe$_2$TiO$_5$ pellet ceramics with various sintering condition is shown in Figure 1 the profiles are similar. The XRD profiles show that the structure of pellets ceramics is orthorhombic after being compared to the XRD standard profile of Fe$_2$TiO$_5$ from JCPDS No.01-070-2728. 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. The XRD data indicates that the synthesis of the Fe$_2$TiO$_5$ pellets has been well prepared from Fe$_3$O$_4$ and TiO$_2$ powder in various sintering condition.
Figure 1. X-ray diffraction patterns of Fe$_2$TiO$_5$ ceramic sintered at: (A) 1200°C for 2 hours in air, (B) 1300°C for 2 hours in air, (C) 1200°C for 8 hours in air, and (D) 1200°C for 2 hours in oxygen gas.

Figure 2. Microstructure of Fe$_2$TiO$_5$ ceramic sintered at: (A) 1200°C for 2 hours in air, (B) 1300°C for 2 hours in air, (C) 1200°C for 8 hour in air, and (D) 1200°C for 2 hours in oxygen gas.
Microstructure of Fe$_2$TiO$_5$ with various sintering condition is shown in Figure 2. Figure 2A and 2B show morphology of Fe$_2$TiO$_5$ ceramics with sintered temperature variation. They clearly exhibit the effect of sintering temperature on the grain size. The grain size becomes larger following the increase of the sintering temperature. This is a consequence of the higher mobility of ions at the higher sintering temperature. The higher the mobility of ions is, the larger the grain growth. The grain size calculated by using of the intercept method is found to be 6.04μm and 26.12μm, respectively. Figure 2A and 2C show morphology of Fe$_2$TiO$_5$ ceramics with sintered time variation. They clearly exhibit the longer of sintering time on the grain size. The grain size becomes larger following the increase of the sintering time. This is a consequence of the higher mobility of ions at the higher sintering time. The higher the mobility of ions is, the larger the grain growth. The grain size calculated by using of the intercept method is found to be 6.04μm and 8.75μm, respectively. Figure 2A and 2D show morphology of Fe$_2$TiO$_5$ ceramics with sintered atmosphere condition. From Figure 2A and 2D are seen that the grain size of the ceramic sintered in oxygen gas is much smaller than that of the ceramic sintered in air. This situation can be explained as follow, relatively poor oxygen for sample sintered in air, make the ceramic could not be well synthesized. Some of Fe$_2$O$_3$ and TiO$_2$ segregated at grain boundaries and inhibited grain growth, producing small grains. The grain size calculated by using of the intercept method is found to be 5.10μm and 6.04μm for sintered in oxygen gas and air, respectively. Sintering atmosphere gases also influence the electrical properties of the pellets Fe$_2$TiO$_5$. In an atmosphere which oxygen-rich grain growth does not work properly causing small grain size. Otherwise, in air atmosphere, grains growth better [17-20].

Electrical characteristics of Fe$_2$TiO$_5$ ceramic with various sintering condition is shown in Table 1 and Figure 3. As shown in Figure 3 and Table 1, it has found that sintering parameter i.e. temperature, time, and atmosphere has reduced thermistor constant, activation energy, and electrical resistance at room temperature and enlarged the grain sizes. Electrical resistance decreasing mechanism occurs through several ways, such as, growing of grain size due to difference of sintering parameter. The larger grain size result a few grain boundaries. The reduced grain boundaries caused by increase of ion mobility at higher temperature and longer time during sintering process will cause scattering and increasing of mean free path. Therefore, electrical resistance of Fe$_2$TiO$_5$ ceramics decreases. The obtained constant thermistor (B) is higher than usual market need (2000 K). It is 4167-6149 K. Generally, the fabricated Fe$_2$TiO$_5$ ceramics in this research are still in market price range. Based on the highest constant thermistor B, Fe$_2$TiO$_5$ ceramic which is the best choice for main component of thermistor, is sintered at 1200°C for 2 hour in oxygen gas.

**Table 1.** Electrical characteristic data of Fe$_2$TiO$_5$ ceramics.

| Temperature/Time/Atmosphere | B  | $E_a$ | $R_{rt}$ |
|-----------------------------|----|-------|----------|
| (°C/hour)                   | (K) | (eV)  | (MΩ)     |
| 1100/2/Air                 | 5964 | 0.51  | 17.40    |
| 1200/2/Air                 | 5879 | 0.50  | 13.20    |
| 1300/2/Air                 | 4167 | 0.35  | 2.53     |
| 1200/8/Air                 | 5770 | 0.49  | 7.44     |
| 1200/2/O$_2$               | 6149 | 0.53  | 26.54    |
Figure 3. Electrical characteristic of Fe$_2$TiO$_5$ ceramic sintered at: (A) 1100°C, 1200°C, and 1300°C for 2 hours in air, (B) 1200°C in air for 2 hours and 8 hours, and (C) 1200°C for 2 hours in air and oxygen gas.

4. Conclusions
Fe$_2$TiO$_5$-based ceramics for NTC thermistor prepared from imported Fe$_2$O$_3$ and TiO$_2$ have been successfully fabricated at 1100-1300°C sintering temperature. XRD data show that Fe$_2$TiO$_5$ ceramics sintered in all sintering parameter (temperature, time, and atmosphere) have an orthorhombic crystal structure. From morphology data of Fe$_2$TiO$_5$ ceramics, it has been investigated that higher temperature, longer time, and change of sintering atmosphere lead to decrease of thermistor constant (B), activation energy ($E_a$), and electrical resistance at room temperature ($R_{RT}$). Electrical characteristic data show that Fe$_2$TiO$_5$ ceramics have good electrical characteristic for NTC thermistor purpose with thermistor constant (B = 4167-6149 K) and electrical resistivity at room temperature ($R_{RT} = 2.53-26.54$ MΩ). The values of B and RRT have met a market demand. Sintering temperature, sintering time, and sintering atmosphere is the parameter that can be used to control electrical characteristic of Fe$_2$TiO$_5$ ceramic as required.

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