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VẬT LÝ CHẤT RẮN VÀ KHOA HỌC VẬT LIỆU TOÀN QUỐC LẦN THỨ XI

TUYỂN TẬP BÁO CÁO

Quyển 1

NHÀ XUẤT BẢN BÁCH KHOA HÀ NỘI

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Sách không bán

EFFECTS OF SINTERING TEMPERATURE ON PHYSICAL PROPERTIES OF $(K_{0.5}Na_{0.5})NbO_3$ CERAMICS

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Abstract:

This work reports the synthesis of $K_{0.5}Na_{0.5}NbO_3$ (KNN) ceramics using following raw materials: titanium dioxide, niobium dioxide, potassium carbonate and sodium carbonate by an improved conventional method. The influence of sintering temperatures of the manufactured ceramics and the piezoelectric, ferroelectric properties of these ceramics was studied. It was found that density increased greatly within a narrow temperature range, and got the highest value at 4.2 g/cm^3 at sintered temperature 1090 ^{o}C. However, when the sintering temperature slightly exceeded the optimal one, the density tended to decrease, accompanied by the appearance of abnormal grain growth, which was considered to be due to the intensified volatilization of alkali metal oxides. Suface mophology of KNN sintered at 1090 ^{o}C showed the tetragonal grain shape, corresponded to a ceramic density of 4.2 g/cm^3. The $(K_{0.5}Na_{0.5})NbO_3$ system sintered at 1090 ^{o}C shows the highest values of electromechanical coupling factor/k_p of 0.22 and d_{33} of 61 (pC/N) related to homogenous grains size of orthohrombic phase.

Keywords*:* lead-free ceramic, KNN, sintering temperature.

INTRODUCTION

In previous decades, piezoelectric ceramic systems basing on titanium zirconate lead (PZT) are widely studied and applied in many fields[1-3]. However, due to the toxicity of lead [4, 6], the development of lead-free piezoelectric ceramic systems with excellent electric and piezoelectric properties to replace lead based ceramics in different devices is necessary [6].

Among lead-free systems, ceramics based on (Na, K)NbO_3 (KNN) have attracted a lot of attention due to their prominent ferroelectric properties, high Curie temperature (about 420^{o}C) and environmentally friendly, capable of replacing the lead based ceramics. However, most recent studies have concentrated on the development of KNN-based lead-free ceramics with enhanced piezoelectric properties through doping or texture control. [5,14–17] Although some studies investigated the normal sintering of KNN ceramics with sintering aids such as CuO, and BaO [18–20] still less attention has been paid to the optimization of the normal sintering process of KNN-based ceramics and the relationship among the densification degree and microstructure as well as electrical properties of KNN based ceramics, in spite of its importance. Based on the above considerations, the present work was conducted to investigate the optimal condition of normal sintering of KNN-based ceramics and the influence of sintering condition on the ferroelectric, piezoelectric, and dielectric properties of the resultant KNN-based ceramic samples.

In this paper, ceramic with the chemical formula $(K_{0.5}Na_{0.5})NbO_3$ was made using conventional solid state reaction method. The changes in structural characteristics, dielectric and piezoelectric properties of fabricated ceramics when sintering temperature varied have been carried out./Results of this paper hope to contribute the understanding about microstructure and physical properties of KNN ceramics depending on the change sintering temperature.

EXPERIMENTAL

The general formula of the studied material was $(K_{0.5}Na_{0.5})NbO_3$, with following powders (purity $\geqslant$ 99 %) of K_2CO_3, Na_2CO_3, Nb_2O_5, used as raw materials. Before being weighed, the K_2CO_3 and Na_2CO_3 powders were dried in an oven at 200°C for 2 h to minimize the effect of moisture. Mixed powder was milled in 8 hours to obtain an appropriate distribution of the particle size, the small particle size will improve the reactivity of the powders [13]. Two calcinations at temperature 850°C for 2 h were then performed to obtain compositionally homogeneous powders and the single phase formation [14]. Thereafter the calcined, powder was ball milled again for 16h to create a more uniform distribution of powders and the reaction occurs completely [14]. The ground materials were pressed into disk 12 mm in diameter and 1.5 mm in thick under 100 MPa. In order to limit the evaporation of alkaline elements, these pellets were covered by the powders with the same composition and then were sintered in a sealed alumina crucible at different temperatures for 2 h.

The crystal structure of the sintered samples was examined by X-ray diffraction with CuKα radiation, λ = 0.15406nm (XRD, D8 ADVANCE-Bruker). The grain morphology of the samples was examined by scanning electron microscopy (Novanano SEM 450-Fei). Archimedes method was used to determine sample densities. After covering two faces of each sample by Ag electrode, ceramics were poled in a silicone oil bath at 80°C by applying dc field of 30kV/cm for 20min then cooling under the same electric field. They were aged for 24 h prior to testing.

The piezoelectric properties were determined from the resonance and antiresonance frequency by using an Impedance Analyzer HP 1493A and RLC HIOKI 3532. The dielectric constant at room temperature was calculated by measuring the capacitance C of samples using RLC HIOKI 3532 at frequency of 1kHz.

RESULTS AND DISCUSSION

Figure 1(a) shows the density of the $(K_{0.5}Na_{0.5})NbO_3$ ceramic samples sintered at following temperature 1070°C, 1080°C, 1090°C, 1100°C, and 1110°C. In sintering temperature range of 1070 to 1110˚C, initially, the density of the KNN ceramic increased with the raising of sintering temperature. Ceramic density reached the highest value at 4.2 g/cm^3 corresponding to sintering temperature of 1090˚C. However, it tended to decrease when the temperature was futher increased. At 1110 ˚C sintering temperatur, density got the value at 3.9 g/cm^3 accompanied by the appearance of many pores on warped sample surfaces. It likely due to the evaporation rate of sodium and potassium ions rapidly increase in exceed optimal sintering temperature region./This result is consistent with Margaret's assumption/that when the sintering temperature increases, the Na^+ and K^+ ions at position A in the perovskite structure are lost due to the evaporation. As a result, the number of vacancy in position A in the lattice increases [16].

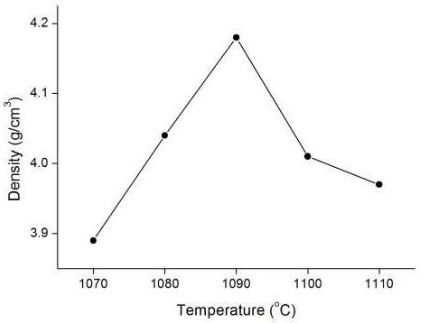

Figure 1. The sintering temperature dependence of the density of $(K_{0.5}Na_{0.5})NbO_3$ ceramics measured at room.

Figure 2 shows the XRD pattern at room temperature with the 2θ range from 20° to 80 ° of $(K_{0.5}Na_{0.5})NbO_3$ sample sintered at 1090°C. It can be seen that samples sintered at 1090°C exhibit single phase structure of perovskite ABO_3, without trace of secondary phases./In order to distinguish whether the fabricated samples have orthorhombic or tetragonal structure, the intensities of the double peaks in the X-ray diffraction pattern are further considered. The corresponding XRD patterns characterized by the double peaks in which intensity of (110) and (220) peaks higher than that of (001) and (020) peaks respectively. According to Skidmore and co-author report, this indicated that all ceramic samples have orthorhombic phase [15].

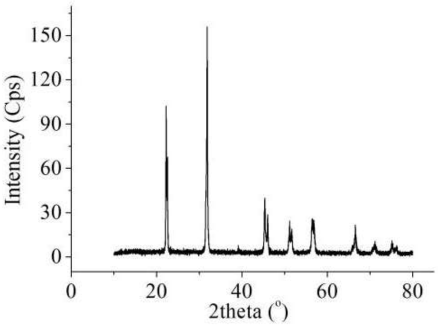

Figure 2. X ray diffraction patterns of the $(K_{0.5}Na_{0.5})NbO_3$ ceramics.

Figure 3 shows the scanning electron microscopy (SEM) micrographs of $(K_{0.5}Na_{0.5})NbO_3$ ceramic sintered at (a) 1070 °C, (b) 1080 °C, (c) 1090 °C, and (d) 1100 °C./

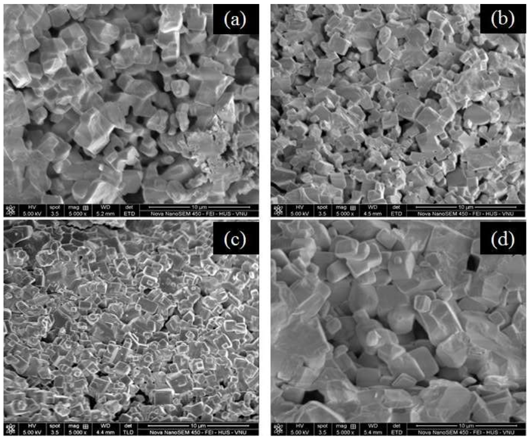

Figure 3. SEM micrographs of $(K_{0.5}Na_{0.5})NbO_3$ ceramic sintered at (a) 1070 °C, (b) 1080 °C, (c) 1090 °C, and (d) 1100 °C.

As shown in Figure 3. (a) and (b), the microstructures of KNN ceramic sintered at 1070 and 1080 °C are not really uniform, the shape of the grains is heterogeneous with the existence of/pores. When the sintering temperature increases to 1090 ° C, the shape of grains becomes homogeneous, its surface is in tetragonal or square form correspond to the highest value of ceramic density. However, when the sintering temperature increases further, some grains tend to grow much larger. As a result, the microstructure sizes of all grains became uneven.

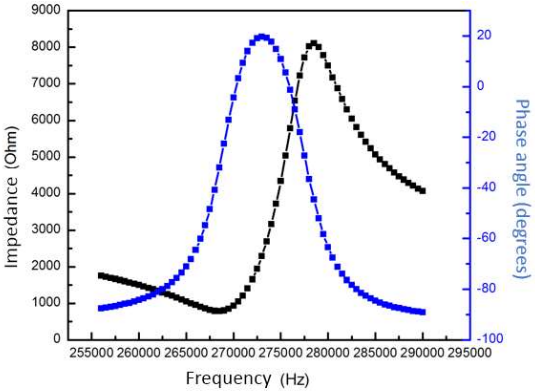

Figure 4. Spectrum of radial resonance of $(K_{0.5}Na_{0.5})NbO_3$ ceramics sintered at 1090°C.

To determine piezoelectric properties of $(K_{0.5}Na_{0.5})NbO_3$ ceramics sintered at 1090 °C, resonant vibration spectra of samples were measured at room temperature (Fig. 4). From these resonant spectra, piezoelectric parameters of ceramic were determined.

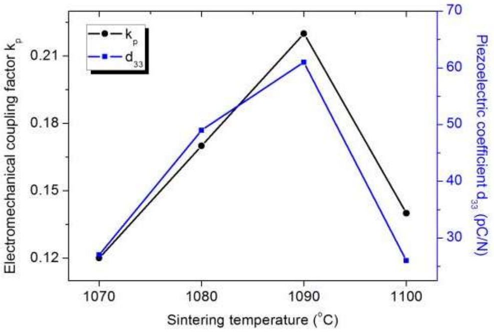

Figure 5. The sintering temperature dependence of the values k_p and d_{33} of $(K_{0.5}Na_{0.5})NbO_3$ ceramics.

Figure 5 shows the electromechanical coupling factors of radial vibration mode (k_p) and piezoelectric coefficient (d_{33}) change as a function of the sintering temperature. The piezoelectric parameters of KNN tend to enhance with the increasing of sintering temperature. The largest values for k_p of 0.22, d_{33} of 61 (pC/N), reached at sintering temperature of 1090 °C. And then piezoelectric parameters rapidly decrease when sintering temperature raising more. These properties of $(K_{0.5}Na_{0.5})NbO_3$ ceramic sintered at 1090 °C corresponding to microstructures of well-faceted and homogeneously distributed grains.

CONCLUSION

The $(K_{0.5}Na_{0.5})NbO_3$ lead-free ceramics were fabricated by conventional solid state reaction. It has been found that sintering densification occurs within a narrow temperature range, and the density decreases apparently when the sintering temperature slightly exceeds the optimal one. Abnormal grain growth tends to occur after the densification is finished and intensified with increasing temperature, which is caused by the volatilization of alkali components.

The highest piezoelectric coefficient d_{33} value of 61 pC/N and electromechanical coupling factor k_p value of 0.22 were obtained for the KNN ceramics sintered at 1090 °C, which is corresponding to the highest density.

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