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Enhancement of $K_{0.5}Na_{0.5}NbO_3$ (KNN) Properties through Optimization of Pressure Applied during Sintering Process via HIP Methods

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ABSTRACT. KNN ceramics were synthesize by solid state reaction method. This investigation were focused on the influence of pressure applied during sintering process via HIP technique to the phase formation, microstructure, density, piezoelectric and dielectric properties of KNN where argon gas pressure applied were varied (2900, 7257 and 14504 psi, respectively). The increment of pressure caused broadening of peak of XRD pattern, increase in peak intensity, density, grain size, and dielectric properties up to 7257 psi. However, at 14504 psi, the grain size start to drop and slightly decrease in dielectric properties.

Keywords: *KNN, HIP, Piezoelectric, Dielectric;*

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1. INTRODUCTION

Lead Zirconate Titanate (PZT) was being used frequently as piezoelectric ceramics as actuators and sensors because of their superior piezoelectric and electrical properties [1]. But, the lead based ceramics will cause a serious environment issue because of their high toxicity of lead and easily to vapourize when exposed to a high sintering temperature [2]. Several works have been done by other researcher in order to develop lead free piezoelectric ceramics with excellent piezoelectric properties besides replacing the lead based materials in various application. There are several example of lead free piezoelectric ceramic such as $Bi_{0.5}Na_{0.5}TiO_3$-based materials, Tungsten bronze type materials and alkaline niobate based materials have been investigated.

Among them, perovskite based lead-free alkali niobates, potassium sodium niobate (KNN) have attracted much attention. $K_{0.5}Na_{0.5}NbO_3$ (KNN) is considered as one of the most promising candidates for lead free piezoelectric ceramics because of its very high Curie temperature (above 400 °C), good ferroelectric properties (Pr = 33 μC/cm^2) and large electromechanical coupling factors [3]. The major drawbacks associated with KNN lies in its poor densification and volatilization of Na which results in non-stoichiometry of the composition thus changing the morphotropic phase boundary (MPB). The addition of excess alkaline oxides to compensate the loss leads to hygroscopic nature of the samples [4]. A numbers of studies have been carried out to improve the properties of KNN ceramics includes the formation of solid solution KNN with other ferroelectric or non-ferroelectric, e.g., KNN-$LiNbO_3$, KNN-$BaTiO_3$, KNN-$SrTiO_3$ as well as the use of sintering aids.

In order to improve the density and piezoelectric of KNN ceramics, advance processing method like hot isostatic pressing have been used in order to established the ~99% density and d_{33} as high as 150 pC/N like reported by [5]. In this research, the pressure of argon gas applied during sintering

process the samples using HIP method have been varying in order to determine the most suitable pressure should be used in order to achieve the highest density body of KNN sample.

2. MATERIALS AND METHODS

$K_{0.5}Na_{0.5}NbO_3$ was synthesized via solid state reaction method using hot isostatic pressing (HIP) technique. A mixture of K_2CO_3 (Merck, 99%), Na_2CO_3 (Merck, 99.9%), and Nb_2O_5 (Merck, 99%) were used as starting raw materials. The raw materials were dried at 200°C for 1 hour in an oven in order to remove any moisture due to hygroscopic nature of the powders. The raw materials were then weighed and mixed in a ball mill using ZrO_2 balls in ethanol medium for 24 hours, then dried and calcined at 850 °C for 4 hours with a heating rate of 3 °C/min. The calcined powder was ground and mixed with 6 wt.% of polyvinyl alcohol (PVA) binder solution before pressed into disks with 13 mm diameter and 2.5 mm thickness at 300 MPa. The pellets were finally sintered by using Hot Isostatic Pressing technique on three different pressure of argon gas applied (2900, 7252 and 14504 psi) at 1080 °C for 30 min. The KNN samples have been labelled as KNN2900, KNN7252 and KNN1450, respectively. The densities of the sintered pellets were measured using gas pycnometer (Micromeritics Accupyc II 1340). X-Ray diffraction (XRD) (Bruker AXS D8 Advance) analysis was carried out to check the phase composition and structure. The microstructure of the samples was observed by using Field Emission Scanning Electron Microscope (FESEM) (Zeiss SUPRA 35VP). The samples were coated by using silver paste on both sides, and then heat treated at 143 °C for 30 min for the dielectric measurement. The relative permittivity (ε_r) and tangent loss (tan δ) have been measured using impedance analyzer (RF Impedance/Material Analyzer 4291B Hewlett Packard) from 1 MHz to 1 GHz.

3. RESULTS AND DISCUSSION

Fig. 1 shows the XRD pattern of KNN sample sintered by hot isostatic pressing technique and being applied at three different pressure of argon gas. In all samples, it shows an identical peak position and peak shape besides no secondary phase is observed. The cells possessed orthorhombic symmetry which is similar to our previous reported [6]. Besides that, the XRD patterns show that as the pressure of argon gas applied increase, the peak intensity becomes larger also exhibits little bit broadening. This is due to average particle size effect as reported by Ghosh et al. [7] and Theivasanthi et al. [8] where the peaks broaden and the widths becomes larger as the particle size become smaller.

Fig. 2 shows FESEM micrograph of KNN pallets sintered at 1080 °C for 30 min using HIP technique on three different pressure of argon gas that applied during sintering process. HIP technique improved the sinterability of KNN ceramic body where creating liquid phase during sintering process which the particles hold strongly between each others. The statement can be support from the fracture surface image which shows that there are many fracture formed on the grains of the KNN fracture sample compared to fracture occur on the grain boundary. On the other hand, increasing the argon gas pressure will improve the relative density of KNN sample near 99% and hence will improve the electrical properties of KNN sample.argon gas applied (2900, 7257 and 14504 psi) via hot isostatic pressing technique as the pressure during sintering was increased, the grain microstructure become finer compared to conventional sintering similar as reported by Egerton and Jaeger [5]. The grain size of sample been applied pressure 14504 psi are substantially smaller than those of lower pressure applied which is 2900 psi due to HIP technique can consolidate ceramic powders into highly dense microstructure without inducing exaggerated grain growth [9].

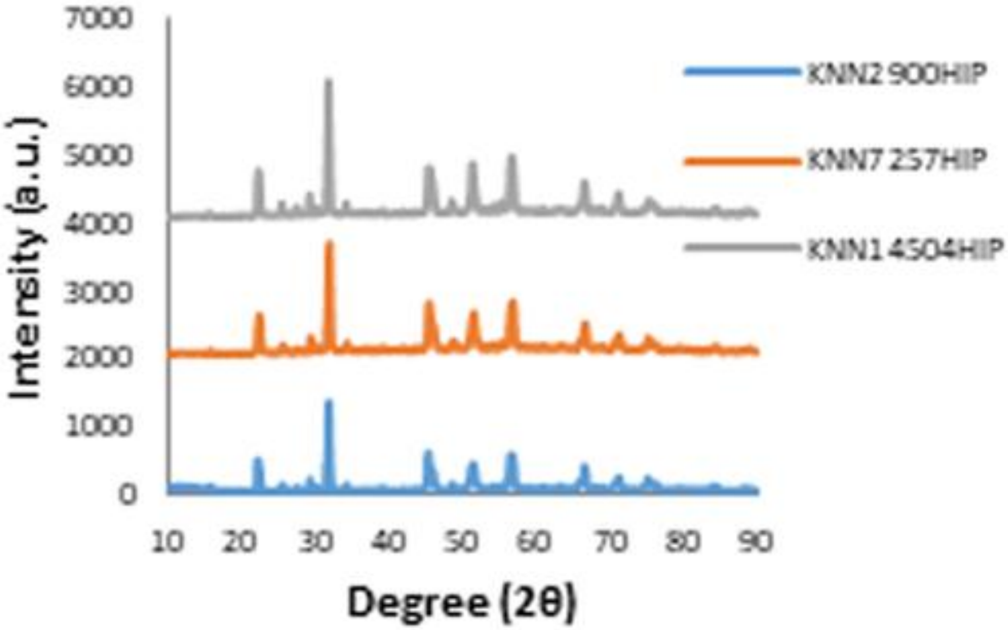

Fig.1 The XRD pattern of the KKNN samples after prepared under different sintering pressure of

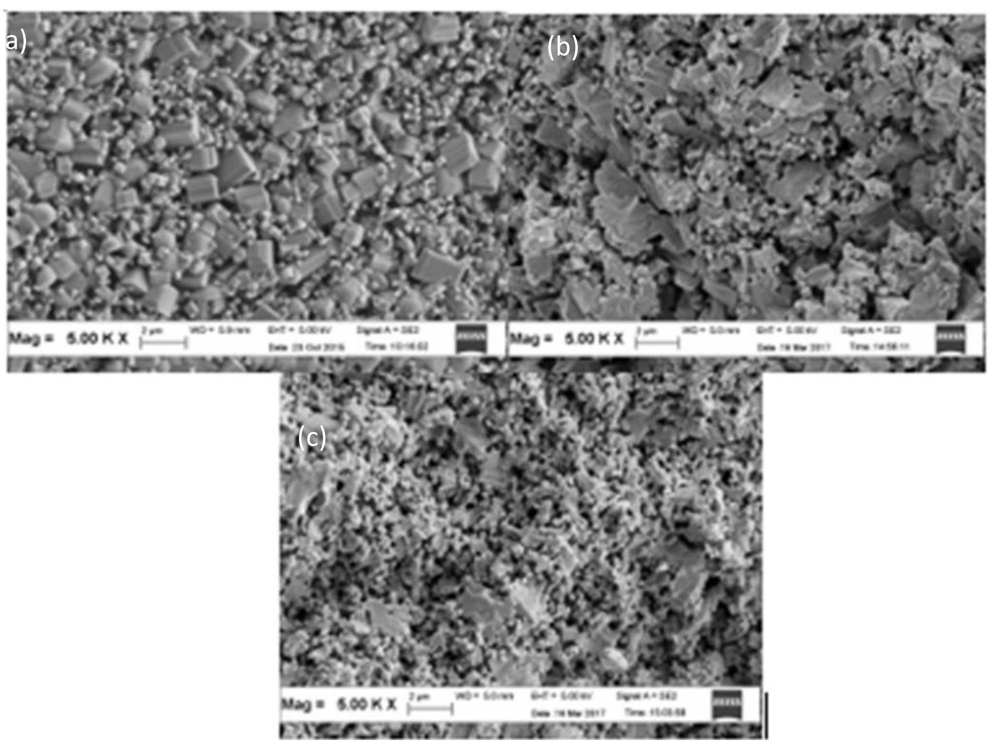

Fig. 2 The FESEM micrograph of KNN samples after applied 3 different pressure during sintering process by HIP (a) 2900, (b) 5257 and (c)14504 psi

KNN exhibits good densification behavior and cuboid morphology was observed under hot isostatic pressure sintering condition which typical for KNN-based ceramic. The density results for KNN samples are shown in Table 1. It can be seen that with increasing the pressure of argon gas during sintering process, the densification of sample improves.

Table 1 Densities of KNN samples fabricated under different pressure of argon gas during sintering process by HIP technique.

Sample	Density (g/cm^3)
KNN 2900	4.4850
KNN 7252	4.5011
KNN 14504	4.6102

Fig. 3 shows the result of dielectric permittivity (ε_r) for KNN sample. The KNN 7252 sample experience the highest ε_r properties compared to the others sample. The ε_r increase as the density increase. We previously reported [6] that dielectric permittivity value will be increased as density of the sample increased. It is due to more space for holding larger quantity of charge at longer period of time [10-13]. Therefore, sample sintered at highest pressure (14705 psi) shows a little bit decrease of dielectric permittivity value due to decreasing in grain size. The decreasing in grain size make the grain difficult to spin during poling process due to pinning effect of the small grain.

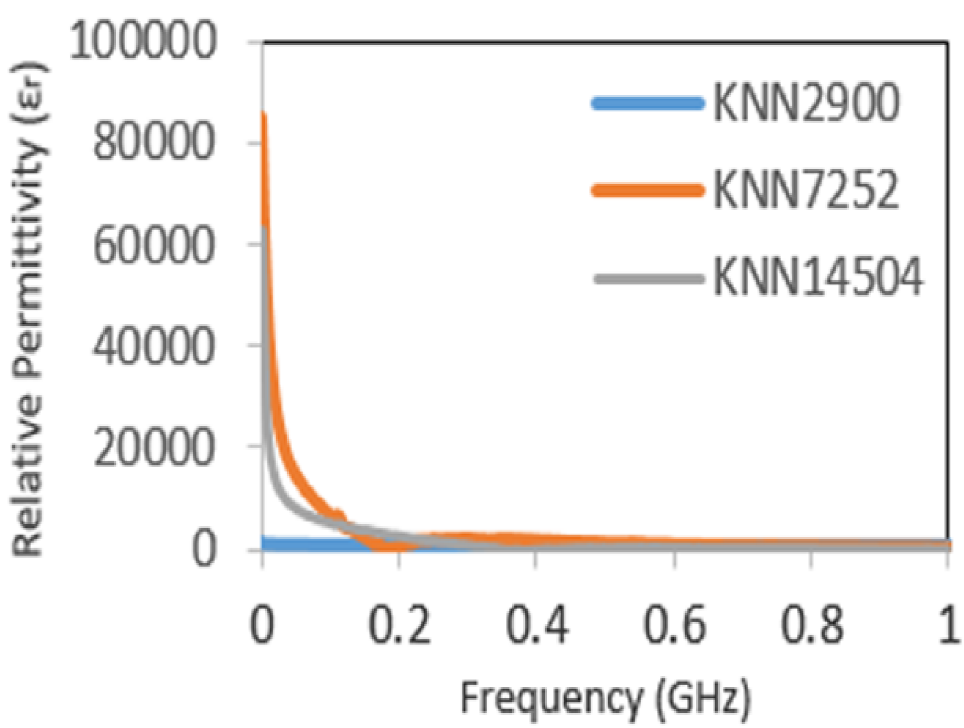

Fig. 3 The relative permittivity of KNN samples after applied 3 different pressure during sintering process (2900, 5257 and 14504 psi)

Fig. 4 shown the variations of piezoelectric properties (d_{33} and k_p value) with pressure applied during sintering process for the KNN sample. The observed d_{33} increases significantly from 155 to 183 pC/N as pressure arises from 2900 to 7257 psi, then decreases at pressure 14505 psi. The observed k_p value also shows significant increase with increasing pressure up to 7257 psi, giving maximum values (0.523) before reduce at 14504 psi (0.4296). The improvement in densification [Fig. 2 and Table 2] may lead to the enhancement in piezoelectric properties [14]. However, at the highest pressure applied (14504 psi), the grain size start to reduce. So, it will disturb the domain

from align easily during poling process which lead to the difficulties to align the domain which cause the reducing in piezoelectric properties of sample KNN 14504 psi [6].

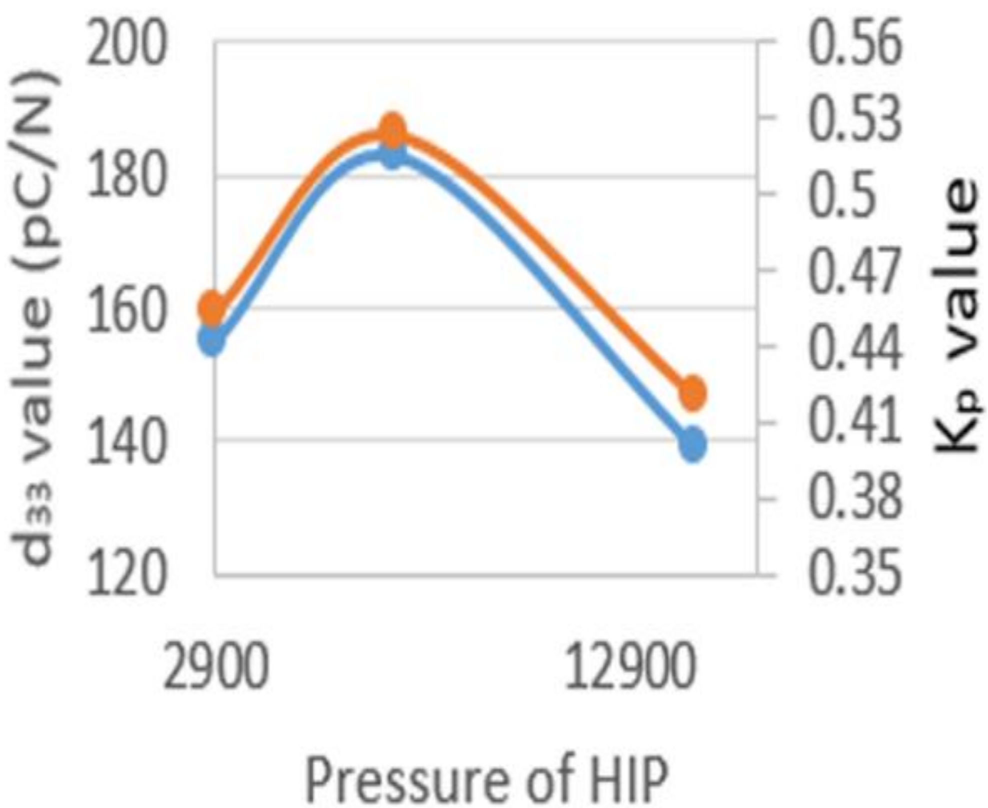

Fig. 4 The variations of piezoelectric properties (d_{33} and k_p value) of KNN samples after being applied 3 different pressure during sintering process by HIP technique (2900, 5257 and 14504 psi)

4. SUMMARY

The effect of different pressure of argon gas applied during sintering process using HIP technique on the phases, structure, microstructure, density and dielectric properties of KNN ceramics was investigated. The increment of pressure caused broadening of peak, increase in peak intensity, density, grain size, and dielectric properties up to 7257 psi. However, at 14504 psi, the grain sizes start to drop and reduce the dielectric properties.

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