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Dielectric Properties of $\text{Pb}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3$ (PFN) Material Synthesized by B-site Precursor Method

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Abstract: B-site precursor method was employed for the synthesis of $\text{Pb}(\text{Fe}_{0.5}\text{Nb}_{0.5})\text{O}_3$ (PFN) Ceramic material. Formation of Perovskite was confirmed by XRD. Study of electrical and dielectric properties were carried out. The increase in dielectric loss and decrease in ac resistivity was observed at higher temperature.

Keywords: Perovskite, XRD, B-site precursor, AC conductivity, Dielectric constant.

I. INTRODUCTION

Relaxor ferroelectrics are a class of ferroelectrics that have a diffuse permittivity maximum, which is frequency- dependent [1,2]. For many practical applications, a very broad permittivity maximum is desirable. Lead iron niobate [$\text{Pb}(\text{Fe}_{1/2}\text{Nb}_{1/2})\text{O}_3$; PFN] is one of the family of lead-based complex perovskites which is of interest as a component in commercial electro ceramic materials. High relative permittivities and low sintering temperatures typically characterize it. Work by Lejeune and Boilot [3] has indicated that PFN is formed indirectly via a sequence of intermediates, some of these being lead niobium oxide pyrochlores. It was also proposed that formation of a perovskite PFN phase requires a high reactivity of ferric oxide, Fe_2O_3 , with other phases in the PbO-NbO_3 system.

Alkaline-earth titanates such as BaTiO_3 , SrTiO_3 , and CaTiO_3 are traditionally used for electronic devices [4]. Because the temperatures required to sinter these materials are generally higher than 1300°C in air [5], expensive noble metals such as palladium, platinum, or their alloys are needed as internal electrode materials. Extensive research has been carried out on dielectric materials that can be sintered at temperatures around 1000°C such that inexpensive electrodes (e.g., silver or silver-palladium) can be utilized [6]. It was reported that perovskite solid solutions based on lead ferroniobate $\text{Pb}(\text{Fe}_{1/2}\text{Nb}_{1/2})\text{O}_3$ are ideal candidates for fabricating multilayer ceramic capacitors with inexpensive electrodes, since they have relatively low sintering temperatures around 900° to 950°C [7]. In addition, these compositions show very high dielectric constants > 20000 providing the possibility for further miniaturization of the multilayer capacitors [8].

II. MEASUREMENT TECHNIQUE

A. Sample Preparation

The B-site precursor method [9] was employed for the preparation of the Lead Iron Niobate (PFN) ceramic studied here. The detailed procedure employed for the synthesis of PFN ceramic is described elsewhere [10]. The samples of PFN ceramics were previously characterized in detail by X-ray diffraction, TG-DTA and SEM[10]. The measured XRD patterns are presented in Fig 1. PFN perovskite formation was confirmed from the XRD pattern.

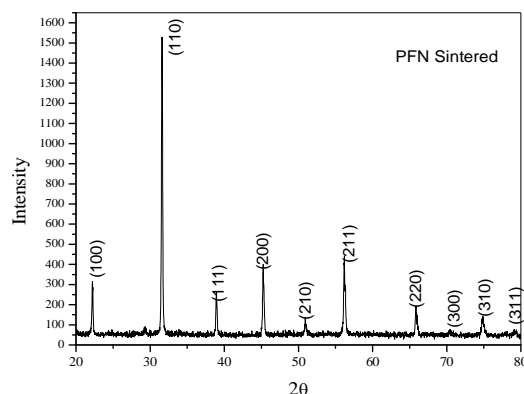


Fig 1. XRD for PFN Perovskite.

B. Dielectric Measurements

For dielectric measurements, the sintered pellets were polished with Al₂O₃ powder (particle size: 1µm) and cleaned using ultrasonic cleaner. The samples were electroded by sputtering gold and then applying air-drying silver paste on the top. By studying their I-V characteristics we could check the ohmic nature of the contacts.

Capacitance and dielectric loss (tan δ) were measured using HP 4194A, impedance/ gain phase analyzer in the frequency range of 100 Hz- 1 MHz. The temperature of the sample was varied from 27°C to 600°C using variable sample holder. The dielectric constant was then calculated from the capacitance and the physical dimensions (area of the electroded portion and the thickness) of the sample. The data was collected during the heating and cooling cycle; however, data collected during heating and cooling cycles were found to match.

III. RESULT AND DISCUSSION

A. Dielectric Properties

The temperature dependence of dielectric constant for PFN at various frequencies are plotted in Fig 2. At low frequency (100Hz), a broad peak can be found at about T_c =118°C. This is in agreement with the literature value. PFN exhibit typical characteristics of diffuse phase transition, which is in agreement with Sun-Gon Jun [11] and N.S. Almodovar et.al.,[12]. The temperature dependence of dielectric loss for PFN at various frequencies is plotted in Fig 3. The dielectric loss increases at higher temperatures.

B. AC Resistivity

The temperature dependence of AC resistivity at various frequencies are plotted in Fig 4. It is clear that, at low frequency a broad peak at 60°C was observed. AC resistivity decreases at higher temperatures and is almost constant. Decrease in AC resistivity indicates the increase in AC conductivity at higher temperatures. This increase in conductivity at higher temperature causes the increase in dielectric loss as confirmed from Fig 4. The values of dielectric constant and dielectric loss for PFN at various frequencies at T_c are summarized in Table 1.

Table No 1. Dielectric constant and dielectric loss for PFN

Frequency	Dielectric constant	Dielectric loss
100 Hz	56063	0.89
1 KHz	35483	0.40
10 KHz	28984	0.10
100 KHz	27088	0.01

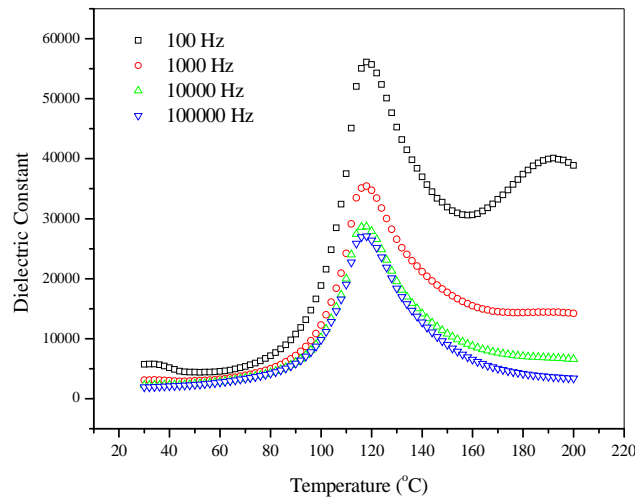


Fig 2. Plot for temperature dependence of dielectric constant at various frequencies for PFN.

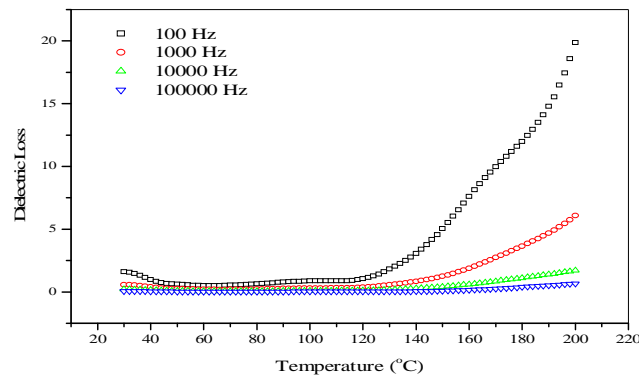


Fig 3. Plot for temperature dependence of dielectric loss at various frequencies for PFN.

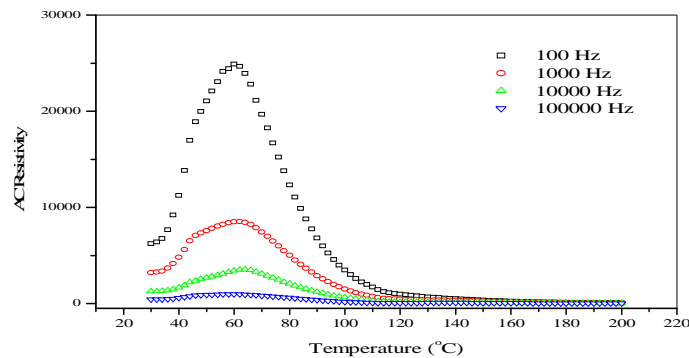


Fig 4. Plot for temperature dependence of AC-Resistivity at various frequencies for PFN.

IV. CONCLUSION

The temperature dependence of dielectric constant showed that the paraelectric to ferroelectric transition in PFN at 118°C is diffusive in character, which is caused by the redistribution of Fe^{3+} and Nb^{5+} ions in B-site of ABO_3 compound. The dielectric loss is observed to increase at higher temperatures. AC resistivity decreases with temperature indicative of an increase in AC conductivity at higher temperatures and is confirmed by high values of dielectric losses at such high temperatures. The possibility of sintering PFN at low temperatures allows the usage of gold and silver paste as electroding material. PFN ceramics are potential materials for ferroelectric memory and high dielectric constant applications.

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