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EFFECT OF ANNEALING ON DIELECTRIC RESPONCE OF $(Pb_{0.8}Ba_{0.2})(Yb_{0.5}Ta_{0.5})O_3$ CERAMIC

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ABSTRACT

Lead barium ytterbium tantalate, $(Pb_{0.8}Ba_{0.2})(Yb_{0.5}Ta_{0.5})O_3$ sample is prepared by conventional solid-state reaction method. To investigate annealing effect, the sample is annealed at 1000 ^oC for 8 hrs in air. A significant change in the dielectric response has been observed for the annealed sample. The dielectric constant increased from 499 to 609 and the dielectric loss tangent decreased from 0.0569 to .0454 at 100 kHz due to annealing. The dielectric peaks of the annealed samples are observed to be more diffused. The dielectric dispersion also increased remarkably for the annealed sample. The annealed sample shows strong relaxor nature.

Keywords: Dielectrics, annealing, diffused phase, relaxor ferroelectrics.

1. INTRODUCTION

Recently, lot of attention has been given to the lead based complex perovskite ferroelectrics having chemical formula $Pb(B' B'')O_3$ due to various industrial applications viz. computer memories, pyroelectric sensors, piezoelectric transducers and multilayer capacitors[1, 2]. In the ferroelectrics family, a major subgroup named relaxor ferroelectrics exhibit high dielectric constant, diffused phase transition and strong dielectric dispersion, have been intensified due to their excellent dielectric and electromechanical properties. $Pb(Yb_{0.5}Ta_{0.5})O_3$ (PYT) is a highly ordered lead based antiferroelectric with two successive phase transitions: (i) a sharp first order phase transition from paraelectric to antiferroelectric around 285 ^oC and (ii) a weak diffused phase transition from antiferroelectric to ferroelectric around 177 ^oC [3]. It is seen from earlier reports that A-site cation substitution causes noticeable changes in the crystal structure as well as dielectric properties. Dai *et al.* [4] reported that by substitution of La in $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) created vacancy in the A-site, that is responsible for the relaxor behaviour. PMN[5] deviates from relaxor nature whereas PZT[6] shows relaxor behaviour with substitution of Ba^{2+} due to compositional fluctuation between Pb^{2+} and Ba^{2+}. In our previous work[7], we have observed that PYT shows weak relaxor behaviour with substitution of Ba^{2+} at the concentration x = 0.2. Therefore, it is interesting to observe the change in the dielectric response of the compound at this particular composition by thermal annealing.

In the present work, the effect of annealing on the dielectric properties of the $(Pb_{0.8}Ba_{0.2})(Yb_{0.5}Ta_{0.5})O_3$ sample has been reported. The variation of dielectric response with the annealing process is studied by taking into account the parameters like dielectric constant (ε_{max}), dielectric loss tangent ($\tan\delta$), ΔT_{max} ($T_{max,\,100\,kHz} - T_{max,,\,1\,kHz}$) and $\Delta T'_{max}$ ($T'_{max,\,100\,kHz} - T'_{max,\,1\,kHz}$) and diffusion coefficient (γ).

2. EXPERIMENTAL PROCEDURE

Polycrystalline sample $(Pb_{0.8}Ba_{0.2})(Yb_{0.5}Ta_{0.5})O_3$ was prepared by the conventional solid state reaction method. The sample was calcined at 950 °C for 2 hrs and sintered at 1300 °C for 2 hrs in PbO atmosphere to avoid Pb loss. The sintered sample is annealed at 1000 °C for 8 hrs in air. X-ray diffraction spectra were recorded for the two samples at room temperature using CuK_{α} radiation. The density of the samples was measured by Archimedes liquid displacement method. The dielectric measurement was carried out over the samples with silver paint applied on both surfaces for electrical contact in the temperature range -100 to 150 °C at low frequencies by using Zentech-1061 LCZ meter.

3. RESULT AND DISCUSSION

3.1.1 X-RAY DIFFRACTION

Fig. 1 shows the room temperature X-ray diffraction pattern of as sintered and annealed $(Pb_{0.8}Ba_{0.2})(Yb_{0.5}Ta_{0.5})O_3$ sample. The X-ray diffraction pattern confirms the single phase nature of both the samples. No appreciable changes are found in the X-ray diffraction pattern for the annealed samples with respect to the as sintered sample. The phase structure is seen to be remaining cubic before and after annealing.

Some fundamental reflections seem to be slightly broadened for the annealed samples, which tentatively mean the reduction in the particle size. As will be seen later, this broadening may be connected to the diffuse phase transition revealed by dielectric measurements. Similar inference has been reported by Kim *et al* [8] for the Ba^{2+} substituted PYN system which is very similar to our system. The density of the as sintered and annealed sample is 9.25 and 9.41 gm/cm^3 respectively. It is obvious that the increase in density will lead to the improvement of electrical properties that observed in our dielectric data discussed in the later part.

3.1.2 DIELECTRIC STUDY

Fig. 2 and 3 show the temperature dependence of dielectric constant and loss tangent of both as sintered and annealed sample respectively at 0.1, 10 and 100 kHz.

It is observed in each case that the dielectric constant (ε) and dielectric loss tangent ($\tan\delta$) increase with increase in temperature, attain maximum values at temperatures T_{max} and T'_{max} respectively and then start decreasing. The maximum value of dielectric constant (ε_{max}) increases remarkably and dielectric loss tangent ($\tan\delta_{max}$) decreases for annealed sample. The increase in dielectric constant may be due to the increase in density and change in microstructure [9, 10 and 11] after thermal annealing. The phase transition modes for the samples evaluated using modified Curie-Weiss law proposed by Uchino and Nomura [12],

$$\frac{1}{\varepsilon'} = \frac{1}{\varepsilon'_{max}} + \frac{(T - T_{max})^{\gamma}}{C} \qquad (1)$$

where γ is the diffuseness exponent and varies between 1 and 2 depending on the nature of the phase transition. The value of γ indicates the increase in diffuseness for the annealed sample. The values of ε'_{max}, $\tan\delta_{max}$, T_{max}, T'_{max} and γ at different frequencies are tabulated in Table I. The value of ε_{max} decreases and T_{max} increases whereas both $\tan\delta_{max}$ and T'_{max} increase with increasing frequency. In each case the diffuseness almost increases with increasing frequency. The range of temperatures, ΔT_{max} and $\Delta T'_{max}$ the set of parameters that can be regarded as indicators of the degree of the relaxor behavior of a particular sample. From the Table I, it is seen that ΔT_{max} and $\Delta T'_{max}$ increases for the annealed sample. The value of ΔT_{max} (10 and 15) and $\Delta T'_{max}$ (10 and 17) for the as sintered and annealed sample indicates an increase in the dielectric dispersion.

The above parameters clearly indicate the increased diffusiveness in the phase transition of all the annealed samples. This typical relaxor behaviour, a manifestation of diffused phase transition, has been observed for the annealed sample.

4. CONCLUSION

The change in dielectric properties of $(Pb_{0.8}Ba_{0.2})(Yb_{0.5}Ta_{0.5})O_3$ sample after thermal annealing has been studied. The single phase of both the samples has been confirmed from X-ray diffraction spectra. The structure of the samples observed to be same for both the samples. The dielectric constant increases whereas the dielectric loss tangent decreases for the annealed sample. The values of γ and ΔT_{max} indicate the increase in diffuseness and dielectric dispersion for the annealed sample compared to the as sintered sample. The variation in dielectric properties of both samples shows a regular trend with increase in frequency. It may be concluded that the sample after annealing is exhibiting stronger relaxor property than the as sintered sample. In our case, annealing is found to be a convenient and economical method to improve the dielectric response.

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TABLE

Table I: Dielectric parameters of as sintered (named as before) and annealed (named as after) sample at different frequencies.

Frequency kHz	ε'_{max}		$\tan\delta_{max}$		T_{max}		T'_{max}		γ	
	before	after	before	after	before	after	before	after	before	after
0.1	548	623	0.046	0.019	7	3	-18	-48	1.15	1.60
1	526	620	0.049	0.034	7	4	-16	-45	1.30	1.62
5	516	617	0.052	0.037	9	7	-14	-40	1.28	--
10	512	615	0.053	0.039	10	9	-12	-38	1.28	1.63
25	506	612	0.055	0.041	11	12	-10	-40	1.32	--
50	500	610	0.057	0.044	12	14	-10	-33	1.32	1.62
100	499	609	0.059	0.045	11	16	-8	-31	1.34	1.75
150	493	606	0.060	0.046	14	17	-8	-30	1.38	1.75
200	491	600	0.062	0.047	16	18	-7	-29	1.39	1.76

FIGURES

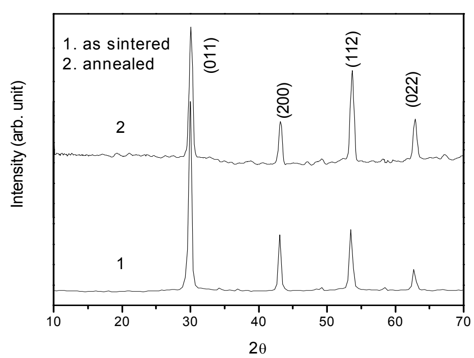

Fig. 1 X-ray diffraction pattern of as sintered and annealed sample.

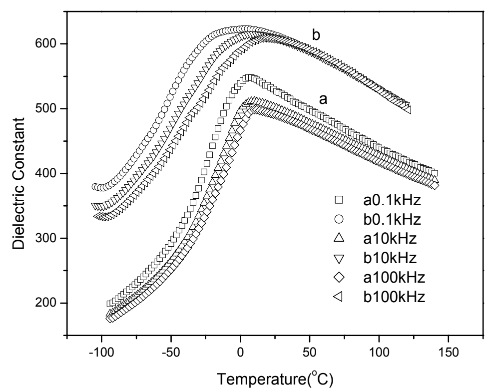

Fig.2 The temperature variation of dielectric constant of (a) as sintered (b) annealed sample at 0.1, 10 and 100 kHz.

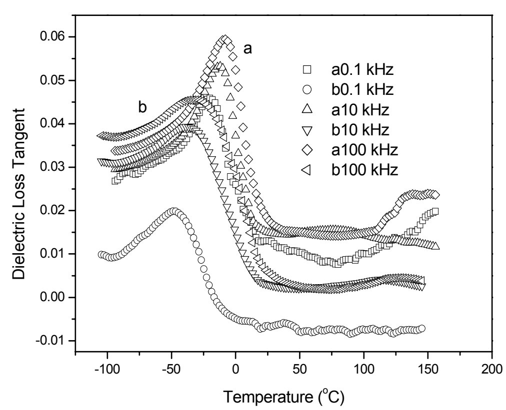

Fig.3 The temperature variation of dielectric loss tangent of (a) as sintered (b) annealed sample at 0.1, 10 and 100 kHz.