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Positron Life-time Studies on Gamma Irradiated Barium Zirconate Ceramic

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Abstract: The zirconates of alkaline earth metals with the general formula $MZrO_3$ (M = Ba, Sr, Ca) normally form perovskite structures and have been projected as potential structural and electronic ceramics. In suitable doped forms they have been claimed to become ionic and/or electronic conductors. However, among the other alkaline earth metal zirconates, barium metazirconate is a value added material, of great importance in the field of technical and electronic ceramics. The present paper deals with the effect of Gamma ray (10kGy) irradiation on the positron lifetime. The BZ ceramic bulk samples (3 mm thick, 10 mm dia.) were synthesized by the ceramic method. The phase of BZ samples were confirmed by comparing their XRD pattern with the standard ICDD pattern No. 74-1299.The grain size is 0.15 micrometer. The gamma irradiation was carried out using gamma chamber 900 with delivered dose of 10kGy available at ISRO Bengaluru. The positron annihilation was done at IGCAR Kalpakam. The life time values are almost same as the reference sample implying that either cationic or neutral vacancies were not formed in BZ due to gamma irradiation. The FTIR measurements were carried out using BOMEM spectrometer. The results show appreciable contraction of Zr-O bond length indicating the effect of hardening in irradiated sample.

Keywords – Barium Zirconate, positron Anihillation, FTIR

1. INTRODUCTION

The zirconates of alkaline earth metals with the general formula $MZrO_3$ (M = Ba, Sr, Ca) normally form perovskite structures and have been projected as potential structural and electronic ceramics. In suitable doped forms they have been claimed to become ionic and/or electronic conductors [1]. However, among the other alkaline earth metal zirconates, barium metazirconate is a value added material, of great importance in the field of technical and electronic ceramics. $BaZrO_3$ is a ceramic oxide of the perovskite family structure with a large lattice constant, high melting point (2893 K), small thermal expansion coefficient, low dielectric loss and low thermal conductivity [1-12]. It is also one of the two parent compounds of the (Pb-free and thus environmental friendly) Ba (Zr, Ti)O_3 solid solutions, which is promising for manufacturing high Q- materials with a variety of applications in microwave industry. Also, the above mentioned properties make it a good candidate to be used as an inert crucible in crystal growth techniques [8, 11], an excellent material for wireless communications [1-2] and a very good substrate in thin film deposition [2-3].

Since ferroelectrics have many interesting applications in memory devices, microwave communications and micro-electro-mechanical devices etc., the study of radiation effects on their physical, structural, electrical and dielectric properties is important for their utility in radiation environment. Irradiation is one of the best methods to study and to change the parameters of the substance. It has been observed that the point defects, extended defects, microstructural and micro compositional changes produced by irradiation can lead to profound changes in properties. Gamma rays are a form of electromagnetic radiation or light emission of frequencies produced by sub-atomic particle interactions, such as electron-positron annihilation or radioactive decay. Gamma rays are generally characterized as electromagnetic radiation having the highest frequency and energy and also the shortest wavelength (below about 10 pm), within the electromagnetic spectrum. Gamma rays consist of high energy photons with energies above about 100 keV.

Positron annihilation spectroscopy is a research tool that is being investigated because it may provide a way to look at defects in a solid matrix. Positron annihilation spectroscopy (PAS) has become a variable tool used to characterize open volume and substitutional defects in functional and other materials[13-17], including ceramics[18-20]. This technique in recent years became an increasingly valuable tool for the study of the electronic and defect structures of materials. The unique aspects of PAS arise from the fact that the positron – electron pair annihilation process, which proceeds by the emission of gamma rays can yield detailed information regarding both the electron density and the electron momenta in the region from which the positron annihilates. There are several attempts to develop a phenomenological model describing the process of positron annihilation some types of $BaTiO_3$[24-26] and $SrTiO_3$perovskites.

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2. EXPERIMENTAL

The required BZ pellets were synthesized by solid state reaction by calicination at 1423 K and sintering at 1723 K. The samples were well crystallized and no parasitic phase was evidenced by XRD. The average grain size was 0.15 µm as measured from SEM micrograph.
The BZ samples were irradiated with gamma radiation (10 kGy). For these irradiated samples, positron lifetime and FTIR measurements were carried out. The gamma irradiation was carried out on bulk BZ samples using Gamma Chamber 900 (delivered dose 10 kGy) available at Indian Space Research Organization (ISRO) Bangalore.
The positron annihilation was carried out to study the lifetime of the positrons. Thereby, the gamma irradiated BZ bulk samples were subjected to positron annihilation to know the nature of defects produced. This was done at IGCAR, Kalpakkam.. When the energetic positron enters solids, gets thermalized, diffuses and ultimately annihilates with an electron. The time spent by the positron in the solid depends on the local electron density around the annihilation site. Measurement of positron lifetime is simpler if one uses Na-22 as positron source. This emits 1.28 MeV γ-ray within few picoseconds of positron emission. The time difference between 1.28 MeV γ-ray emission and the subsequent 511 keV annihilation γ-ray emission is measured over few nanosecond range. Infrared spectroscopy is built around a BOMEM DA-8 FTIR spectrometer that operates over an extended range of 10 - 25000 cm^{-1}. This extended range covering the Far-IR, Mid-IR and the visible region is covered using a set of three sources, beam splitters and detectors.

3. RESULTS AND DISCUSSION

3.1 Charecterization

The phase of BZ samples were confirmed by comparing their XRD pattern with the standard ICDD pattern No. 74-1299 and is shown in Figure 3.1. The XRD patterns match well with the standard ICDD pattern confirming the 100% perovskite phase formation with no trace of any pyrochlore phase. The presence of sharp peaks confirms the high crystallinity of the material. The microstructure analysis of unirradiated BZ was carried out and is shown in Figure 3.2. The average grain size was 0.15 µm.

3.2 Positron life time measurement

The positron lifetime t is a function of the electron density at the annihilation site. The annihilation rate l, which is the reciprocal of the positron lifetime t, is given by the overlap of the positron density, $n_+(\mathbf{r}) = |\Psi^+(\mathbf{r})|^2$ and the electron density $n_-(\mathbf{r})$,

$$\lambda = \frac{1}{\tau} = \pi r_0^2 c \int |\Psi^+(r)|^2 n_-(r)\gamma dr$$

r_0 is the classical electron radius, c the speed of light, and **r** the position vector. The correlation function $\gamma = \gamma[n_-(\mathbf{r})] = 1 + \Delta n_-/n_-$ describes the increase Δn_- in the electron density due to the Coulomb attraction between a positron and an electron. This effect is called enhancement. When positrons are trapped in open-volume defects, such as in vacancies and their agglomerates, the positron lifetime increases with respect to the defect-free sample. This is due to the locally reduced electron density of the defects. Thus, a longer lifetime component, which is a measure of the size of the open volume, appears. The strength of this component, i.e. its intensity, is directly related to the defect concentration. In principle, both items of information, i.e. the kind and concentration of the defect under investigation, can be obtained independently by a single measurement. This is the major advantage of positron lifetime spectroscopy compared with angular correlation of annihilation radiation or Doppler-broadening spectroscopy with respect to defect issues.

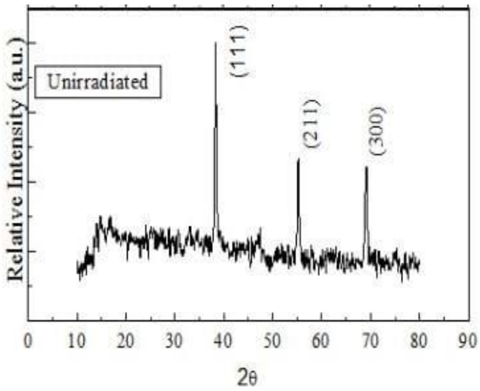

FIGURE 3.1 XRD of Barium Zirconate ceramic

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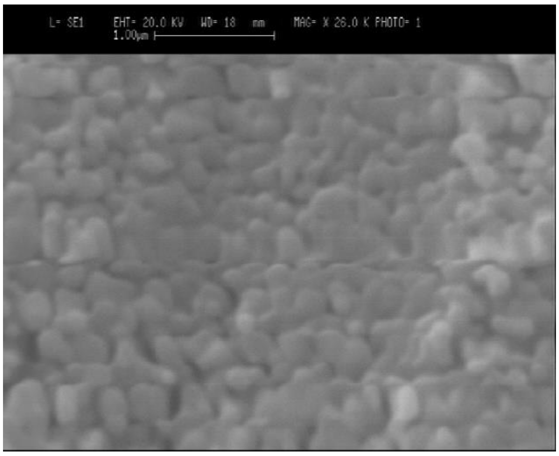

FIGURE 3.2 SEM micrographs of UI, Barium Zirconate

Positron lifetime measurements can be used to study the cationic and neutral vacancies in oxides. Hence, with the aim of studying the cationic and neutral vacancies if any, existing in gamma irradiated BZ, the positron lifetime measurements have been carried out using BaF_2 based positron lifetime spectrometer with a time resolution of 240 ps. The measurements were carried out in reference and gamma irradiated BZ ceramic at room temperature. Measurement in the reference sample shows the existence of only one lifetime component, implying that all the positron annihilate in the defect free bulk of the reference sample. Lifetime in reference sample is 180 ± 2 ps. On the other hand, it is seen that even in the gamma irradiated sample there exists only one lifetime component. This means that the positrons annihilate at a single site with a lifetime of 179 ± 2 ps. These values of lifetime are almost same as that of the reference sample implying that either cationic or neutral vacancies are not formed in BZ due to gamma irradiation for particular energy or if they are formed and are mobile at room temperature leading to the recovery of these defects. In other words, either they are not formed or they do not survive at room temperature. If by any chance there exists only negative ion vacancies i.e., oxygen vacancies in BZ following gamma irradiation, positron could not detect these defects directly. This is due to the reason that the effective charge of oxygen vacancies is positive and positron being positively charged particle cannot detect these defects.

3.3 FTIR measurements

FTIR measurements have been carried out using BOMEM spectrometer. Figure 3.3 and Figure 3.4 shows the FTIR spectra for unirradiated for the evolution of modes in irradiated sample with annealing treated BZ sample.

The isochronal annealing with a step of 20 min was done. FTIR results from literature reveal Zr-O stretching mode around 545 cm^{-1}. In the unirradiated sample itself there seems to be orthorhombic distortion resulting in a distribution in Zr-O bond lengths. But, this builds up appreciably in gamma irradiated sample and with isochronal annealing the spectra become same as that of reference sample for annealing beyond 423 K.

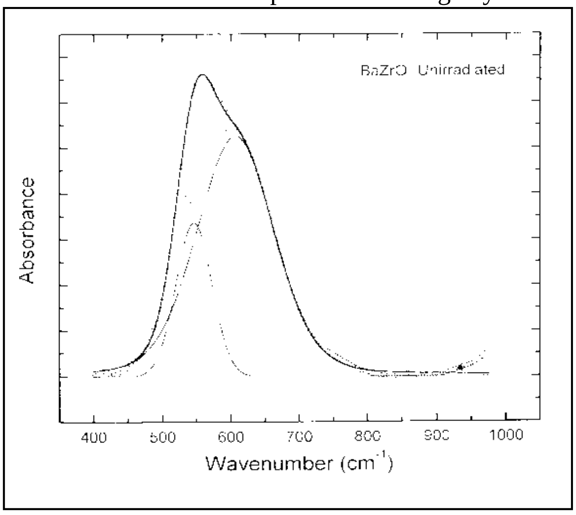

FIGURE 3.3 The FTIR spectra for UI BZ

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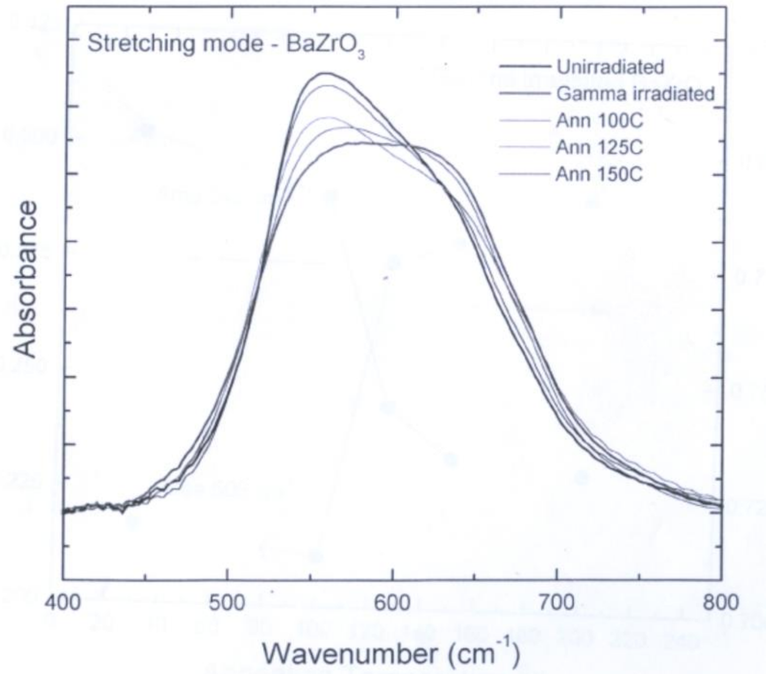

FIGURE 3.4 The FTIR spectra for evolution of modes in irradiated BZ with annealing

This is presumed to be due to increasing lattice distortion in ZrO_6 leading to distribution in bond lengths of Zr-O. There seems to be an appreciable contraction of Zr-O, as the effect of hardening is more in irradiated sample. This effect gets annealed out in reference sample with lesser distortion and lesser distribution in reference sample.

4. CONCLUSION

The results of positron life time and FTIR studies on Barium Zirconate samples prepared by ceramic method are presented. The life time of positrons in the reference sample is 180 ± 2 ps, while it is 179 ± 2 ps for Gamma irradiated sample. This implied that the positron annihilate at a single site. The same life time values in the reference sample and gamma irradiated sample implies either cationic or neutral vacancies were not formed in the BZ sample due to gamma irradiation for particular energy or if they were formed and were mobile at room temperature.

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