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Gamma Radiation Induced Electrical Response of $BaSrTiO_3$ Interdigitated Capacitor

5.1 INTRODUCTION

Electrical characterization of BST thin film is one of prime requirement to integrate BST capacitors in RF and microwave applications. Gamma radiation induced effects in thin film forms an important part of electrical study for BST films. In keeping with the practical aspects, the performance of the device under such working conditions was investigated in the laboratory scale, by applying gamma radiation exposure to the dielectric varactor and observing the change in leakage current and capacitance-voltage characteristics. The results generated through such a study enables one to extrapolate the lifetime of the device under test and further led to improvement of the material properties to be used for such applications. Polarization mechanism relates the changes in dielectric constant across the frequencies and deviation in dielectric loss. The total polarizability is dependent on various mechanisms as discussed in Chapter 2 which includes electronic, dipole, ionic and space charge polarisation. The prime importance of such study is to confirm sustainability of dielectric constant (tunability) and loss tangent on the electric field in RF frequencies region because of their major role for realization of device. Metal oxide thin films are widely used as electronic components for their application in space, defence and nuclear industry for futuristic technologies including memory storage and frequency agile devices. As discussed in previous chapters ferroelectric thin films have become increasingly important as future materials for electronic devices. Metal-oxide-semiconductor device are the key functional elements in micro-electronic field. Miniaturization of the devices requires a reduction of gate oxide thickness where reduction of gate oxide thickness leads to an increase of gate leakage current. Use of dielectric materials as insulating layer was suggested in order to reduce the leakage current. In metal-oxide-semiconductor (MOS) structure, many gate oxides have been investigated to replace SiO_2 by high-k dielectrics. It is well known that the dielectric permittivity can be varied with applied electric field to meet the requirement of modern microwave devices. In recent years, the development of tunable dielectric materials for application of voltage controlled tunable filters and phase shifters received much attention and the tunability is considered as one of the important design parameter [Cole *et al*, *2003*]. Perovskite oxides such as $Ba_xSr_{1-x}TiO_3$ (BST) and $SrTiO_3$ (STO) are being considered prime choice of material systems for microwave devices. BST has been proved as promising material for such application with material properties of low intrinsic carrier generation, high-k dielectrics, high break down field and high charge storage density [Chun *et al,* 2008]. Subramanyam *et al* has reported that the BST is the best suitable among complex oxides thin film for room temperature based voltage tunable dielectric thin films with its low microwave loss and large dielectric tunability to meet the requirements of various electronic device applications [Subramanyam *et al*, 2013].

Barium titanate ($BaTiO_3$) has been found highly resistant to higher dose of gamma radiation [Keating and Murphy, 1969], which favors the pervoskite BST structures could be highly resistant against gamma radiation. BST has grown by several deposition techniques and sputtering is found suitable commercially available technique being as large surface area deposition with better uniformity at lower deposition temperature. MIM configurations have been reported to investigate device electrical properties including C–V and I-V characteristics. Two different types of varactors i.e. MIM and interdigitated capacitor (IDC) have been advised for tunable components and circuits. In general, IDC devices are simpler to fabricate requiring only a single metallization step compared to multiple steps. Another advantage of IDCs is that the thin film can be directly deposited onto the substrate and can therefore be annealed at higher temperatures. The relative dielectric constant of the device can be tuned by the applied electric field between fingers of IDC capacitors. The use of such devices considers the effect of ionizing radiation and concern about the stability of the devices in radiation environment.

Si and SiO_2 thin film based devices have been largely investigated for past several years and found to be sensitive to ionizing radiations. Device deterioration and increased power consumption reduces operation life time of electronic components in space systems. Many researchers have investigated radiation induced defects in semiconductor device [Yas, 2012; Trefilova *et al,* 2001] and reported that irradiation of metal oxide devices with high level of ionizing radiation results in generation of point defects, defect clusters and dislocation center [Srour and Palko, 2013; Arshak and Korostynska, 2001]. The most crucial part of MIM devices are oxide layer where electron hole pairs are generated along with point defects due to irradiation and leads to changes in device properties [Miao *et al*, 2009]. Degradation of carrier mobility and change in carrier concentration has been shown by creating radiation induced defects at sufficiently higher gamma radiation dose. Charge distribution in the devices is significantly influenced by the radiation induced phenomenon which can distort local field near interfaces and defects. Therefore new materials and devices are required to be investigated for their reliability under gamma radiation exposure. Since, much literature is not known about the radiation hardness of these materials and un-doubtfully these efforts will improve the understanding of the radiation damage phenomenon in these new systems. Therefore it is requirement to understand the response of BST based devices under gamma radiation environment for their suitability in high energy ionizing radiation.

In this chapter we attempted to investigate the gamma radiation effects on electrical properties of IDC patterned $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) thin films, deposited on sapphire substrate by RF magnetron sputtering. The IDC structure was fabricated using photolithography technique. Gamma irradiation induced changes of the BST based tunable capacitor have been investigated under various doses from 0 kGy to 600 kGy. Structural and surface morphological studies have been carried out for un-irradiated and irradiated film and revealed that the grain sizes and crystallinity are strongly depended on gamma irradiation doses. The significant radiation induced changes in microstructure have been correlated with macroscopically C-V and leakage current behavior of BST varactor.

5.2 DEPOSITION OF BST ON SAPPHIRE SUBSTRATE

Sapphire, being as insulator, is an ideal substrate for microwave integrated circuit fabrication because of its low dielectric losses and low cost. Electronically tunable capacitors (varactors) were fabricated on top of the BST films by patterning IDC electrode structures. High capacitance tunabilities were achieved for the films grown on sapphire at 30 V bias. Sapphire also has a very high resistivity compared to standard silicon, which means that low loss microwave transmission lines can be realised. The BST film (200 to 300 nm) is deposited, followed by thin metal layers Au(500 nm)/Ti(100 nm). Several other groups have investigated parallel plate BST varactors for microwave applications [Xu *et al*, 2004; Im *et al*, 2000; Jeon *et al*, 2002]. An advantage of IDC device is that the electrode metallisation contributes less to the device losses than in the case of parallel plate capacitor [Tagantsev *et al, 2003*]. The reduction in growth temperature is extremely important for tunable devices fabrication due to material selectivity and thermal sensitivity [Nam *et* al, 2000].

BST films were deposited by RF magnetron sputtering using $Ba_{0.5}Sr_{0.5}TiO_3$ composite target. Fabrication process started with single side polished crystalline r-plane sapphire wafer. Prior to use, wafers were cleaned by standard semiconductor cleaning process using Trichloroethylene, Acetone and Methanol. Cleaned and dried wafer loaded in the process chamber and base vacuum was achieved better than 2×10^{-7} Torr. Film deposition pressure was maintained 15 mTorr in the process chamber.

5.2.1 Electrode Patterning on BST

For electrical characterization at RF and microwave frequencies, IDCs were fabricated on BST/Sapphire wafer using standard photolithography process. The interdigitised structure has wide surface area in comparison to vertical MIM structure and preferred to study the electrical response at high radiation exposure. It offers simplified bias schemes and lower dispersion loss in this configuration which results into a high device quality factor. Interdigitated capacitor's masking steps are shown in Figure 5.1. The BST thin film layer was deposited straight on the substrate using sputtering technique. The top electrode, gold was patterned using lift-off technique to form the interdigitated metal fingers. The IDCs fabrication needed only one mask step and thus highly suitable for low cost devices.

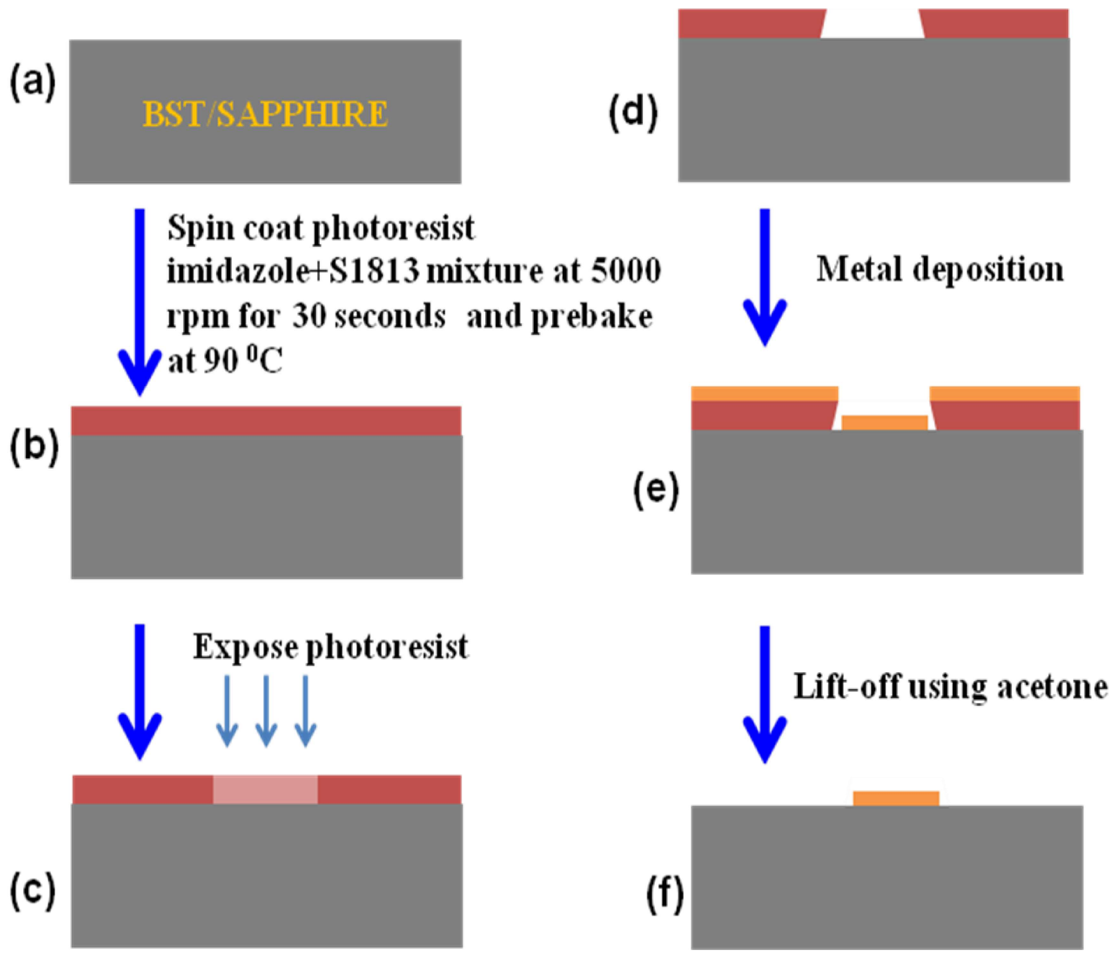

Figure 5.1: IDC patterning steps by photolithography

The removal of bottom electrode prevents microstructural degradation at the dielectric electrode interface. Electrode metallization Ti(30nm)/Au(200nm) were patterned by standard lift-off process. The IDCs composed of 10 fingers with the finger dimensions of 4 µm width, 3 µm spacing and finger length 390 µm as shown in Figure 5.2.

The fabricated IDC devices were irradiated for different gamma exposure in air at a dose rate of 3.4 kGy/hr using Co-60 gamma radiation source. The gamma doses were increased in different steps to the cumulative doses from 0 kGy to 600 kGy. The device was mounted vertically in the gamma chamber, exposed from all sides, realizing the radiation scenario in space and nuclear applications.

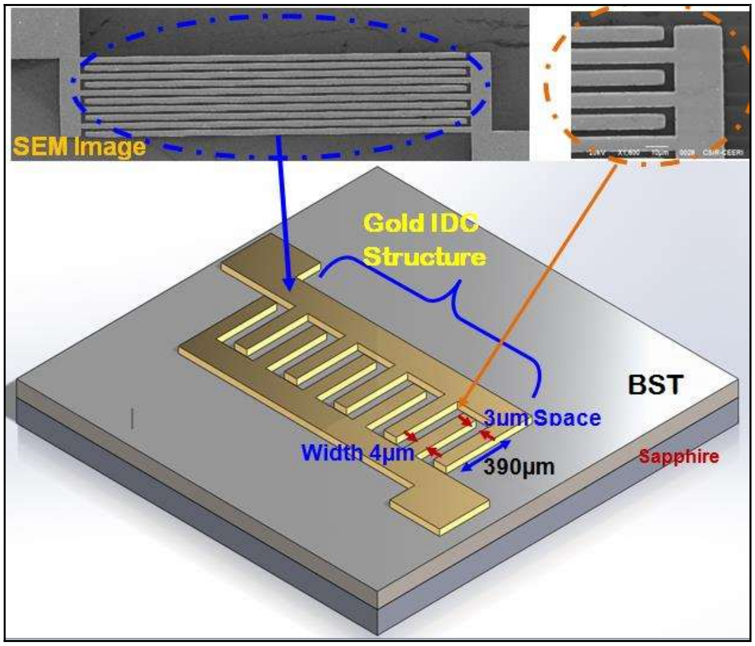

Figure 5.2: Schematic view of BST/Sapphire based Au plated IDC structured Device. SEM image of top view is shown in inset.

5.3 STRUCTURAL AND SURFACE MORPHOLOGICAL CHARACTERIZATIONS

The structural and surface morphological characterizations were carried out using X-Ray Diffractogram (XRD) and Atomic Force Microscope (AFM), before and after gamma irradiation of BST based IDC capacitors. Figure 5.3 shows XRD 2θ-ω scan of BST thin films obtained for different gamma irradiated samples (0 kGy-600 kGy).

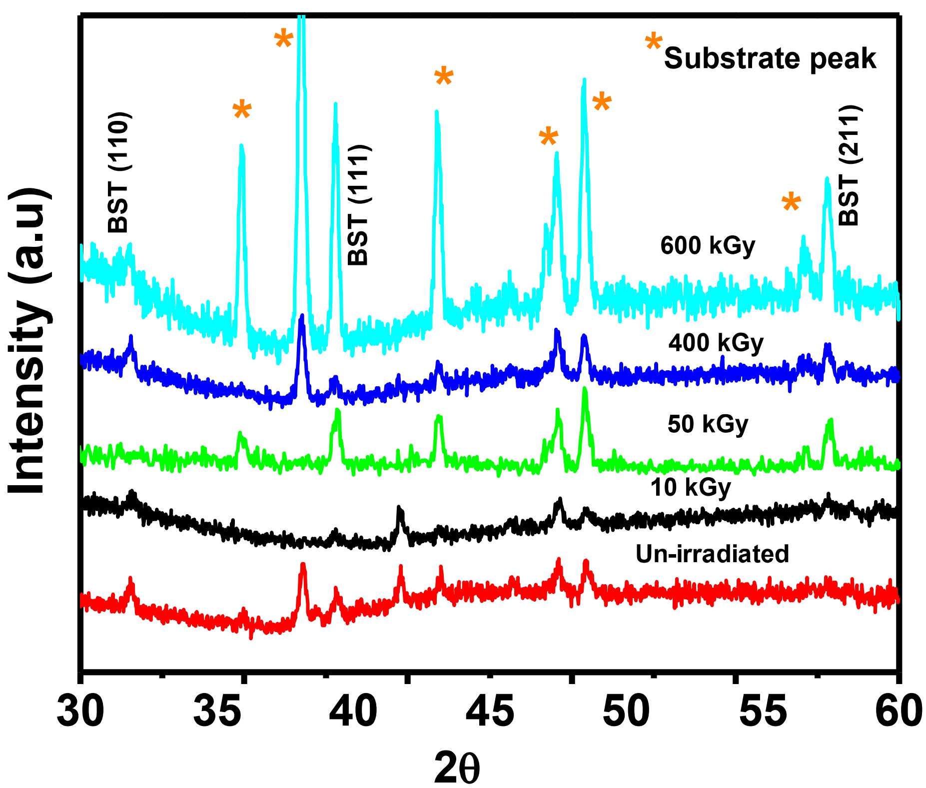

Figure 5.3 : XRD spectra of the Ba0.5Sr0.5TiO3films as a function of gamma dose up to 600kGy

The XRD pattern of BST film shows pervoskite structure with the diffraction peaks of (110), (111) and (211) plane of the deposited film. It is revealed that basic crystal structure remains the same with increasing gamma radiation doses, however a slight change in crystallinity was observed. Radiation induced structural changes has been observed as the gamma rays impinge upon the films, takes place at the high energy disordered boundaries. Effective size of the crystallite is therefore decreased, but larger amorphous regions also exhibit nucleation of new crystallites, resulting in slightly increased overall crystallinity [Wang *et al*, 2002]. The interface between the crystallite boundary layers, which can be simply grain boundaries, is essential to the polarization as seen in other systems [Hagiwara *et al,* 1998].

Atomic force microscopy (AFM) was carried out over an area of 3μm x 3μm on BST after samples irradiation under different gamma doses. The basic morphological changes which accounted for the more visible grain structure as well as the changes in surface roughness were studied. Samples which were irradiated with different gamma doses showed an observable change in the microstructure. The surface morphology is shown in Figure 5.4 for different gamma doses. The surface roughness of BST thin films are observed with an average surface roughness in the range of 2nm to 7nm and average grain size was decreased with increasing gamma radiation dose. Combination of the voids with each other after irradiation, decreases in roughness and grain size due to the excessive decrease in number of voids as similar to reported by Baydogan *et. al* [Baydogan *et al*, 2013].

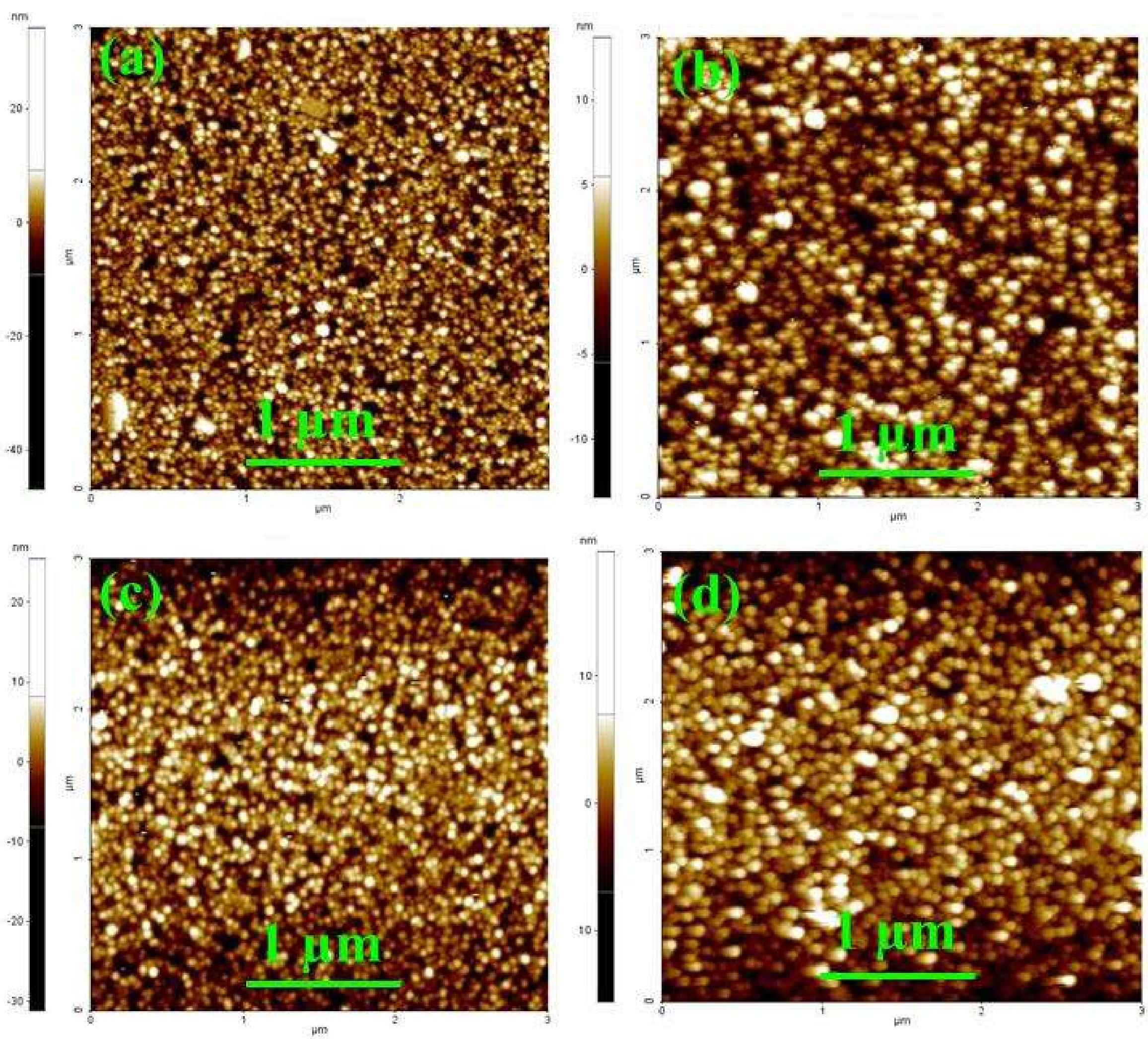

Figure 5.4 : AFM images of BST thin film before and after gamma irradiation, (a) unirradiated, (b) 50 kGy (c) 400 kGy and (d) 600 kGy.

5.4 ELECTRICAL CHARACTERIZATION

Electrical characterization system (Keithley SCS-4200) has been used to study the capacitance–voltage (C–V) and current-voltage (I-V) behavior of the IDC capacitor before and after gamma irradiation for different cumulative doses. The measurements were carried out with a constant time difference after each gamma exposure dose to account for prolonged generation of interface states after irradiation. The C–V curves were measured at different frequencies from 200 kHz to 1 MHz with 30 mV ac signal.

5.4.1 Capacitance – Voltage (C-V) Measurements

Changes in dielectric properties of the device can be measured by C-V characteristics of IDC structured device. The C-V measurements of un-irradiated IDC capacitor were carried out at room temperature for different frequencies as shown in Figure 5.5. DC bias swept has been performed from -30 V to 30 V at a rate of 0.25 V/s. These characteristics follow well known bell shaped behaviour of IDC capacitor structure and attributed by changing the direction of polar molecule depending upon the applied electric field. The capacitance decreases as frequency increases from 200 kHz to 1 MHz. Existence of bulk defects and interface states in the un-irradiated device itself reveals dependence of the capacitance on voltage and frequency both. This frequency dispersion may be attributed with reduced interface barrier height between dielectric film and electrode by these traps, located near the dielectric metal interface.

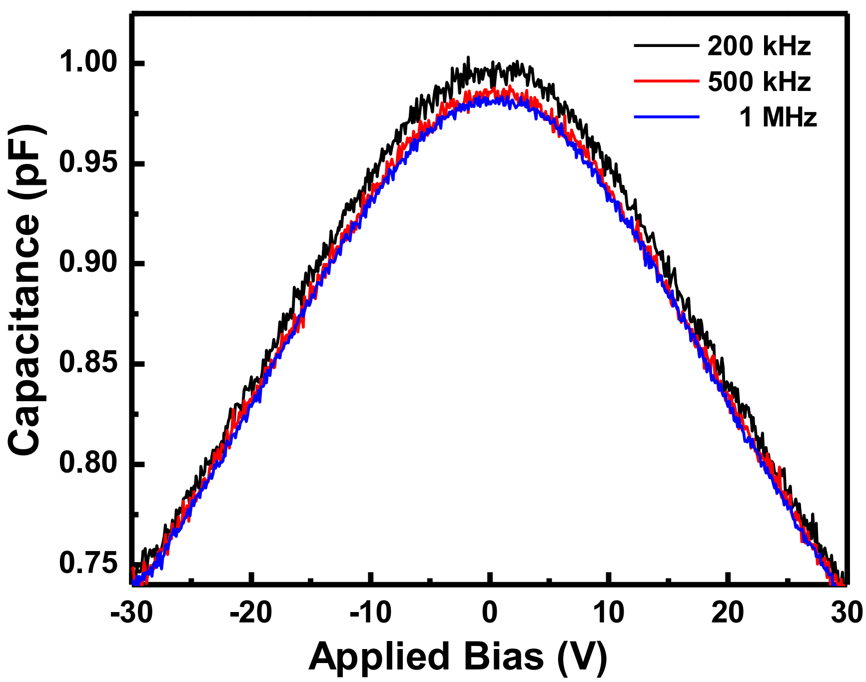

Figure 5.5 : C-V measurements of pristine device at 200 kHz, 500k, and 1 MHz for IDC capacitor

It has been mentioned earlier that the frequency agility in the tunable microwave devices is achieved through the non linear response of ferroelectric thin films to applied bias voltage. It is also known that the tunability is closely related to the soft phonon mode, which originates from the vibration of Ti and O ions in oxygen octahedra with opposite directions in ABO_3 perovskite lattice. If an external force acts upon the system in such a way that the ionic displacement is constrained then the soft phonon frequency and the dielectric constant changes significantly. Field-induced hardening of the soft mode frequency (anharmonic restoring forces on the Ti ion) will lead to a decrease in the static dielectric constant, termed as "Tunability".

Tunability of the devices has been calculated as a function of applied voltage as given in Eq. (5.1).

$$Tunability = \frac{C(0) - C(V)}{C(0)} x100 \qquad (5.1)$$

where C(0) is the maximum capacitance, generally obtained at zero applied bias and C(V) is the capacitance at certain bias voltage when the bias field is applied to the thin film. The observed tunability is around 25% and this is close to earlier reported values for BST IDC at 30V maximum bias voltage [Potrepka *et al*, 2006].

Radiation induced changes are expected while irradiating the device by ^{60}Co gamma-ray source and the radiation damage caused exclusively due to point defects. Since, interactions of Compton electrons having a maximum energy of around 1 MeV are primarily responsible for gamma interaction which does not allow cluster production, as reported by Huhtinen et.al [Huhtinen *et al, 2002*]. We were not having good idea about changes in BST after gamma exposure to the device therefore we started the electrical measurement with low gamma doses. We initially measured the C-V characteristics of BST varactor with varying the dose from 225 Gy to 10 kGy as shown in Figure 5.6 at 500 kHz frequency. It was observed that there was no significant change in the C-V behavior below 10 kGy gamma dose.

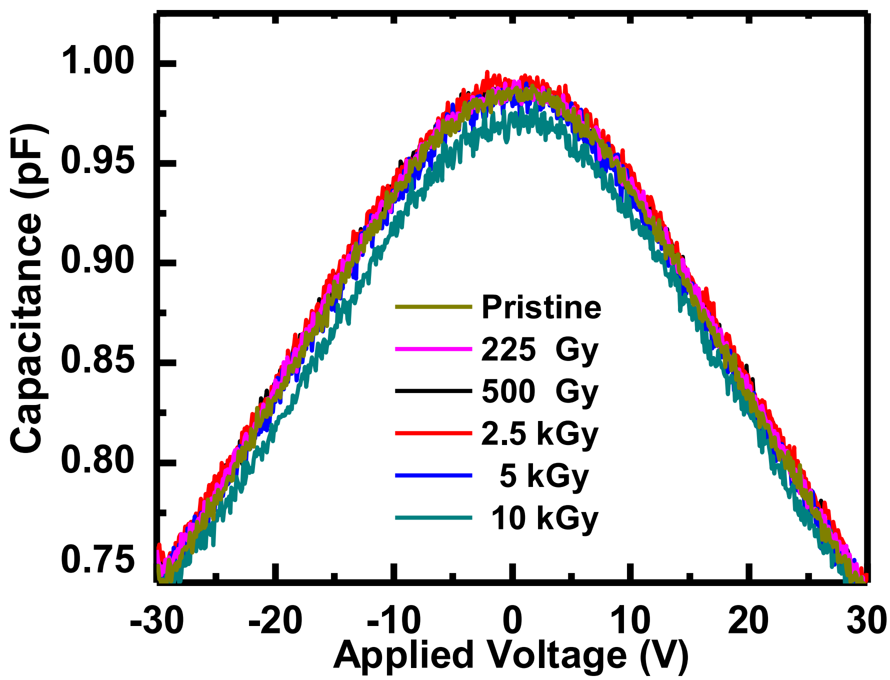

Figure 5.6: Gamma irradiated response of BST IDC capacitor for initial low gamma doses

Hence, further the capacitance of the device was investigated as a function of gamma dose from 10 kGy to 600 kGy at two different frequencies of 500 kHz and 1 MHz as shown in Figure 5.7 and 5.8 respectively. A decrease of the capacitance was observed when the gamma irradiation dose raised from 0 kGy to 50kGy for both the measurement frequency. The capacitance of the IDC structured capacitor device is slightly lower than that of the without irradiated device. It has been shown by Dawber et. al. that Schottky and Poole–Frenkel emissions, ohmic, space-charge limited, Fowler–Nordheim tunneling, hopping conductivity and conductivity via the grain boundaries are the possible conduction mechanisms in ferroelectric films [Dawber *et al,* 2005]. Therefore C-V measurements in BST IDC devices were affected by mobile charge carriers, might have contributed and resulted in drifting inside the grains as well as along grain boundaries in response to the bias field [Schroder, 1998]. The change in the electrical properties of the device can be attributed to the generation of the V_2O center [Huhtinen *et al*, 2002]. The similar observations were also reported by Fretwurst *et al* [Fretwurst *et al*, 2002], rightfully be regarded for the BST thin film IDC structured device by defect analysis for an understanding of macroscopic damage effects. Decrease of the capacitance may be determined by a combination of intrinsic size effect and the "dilution" effect due to the low -permittivity grain boundaries [Frey and Payne, 1996]. The boundary layers are very sensitive to the processing parameters and to the quality of the polycrystalline film. It is largely accepted that grain boundary layer is a highly atomic disordered region, even amorphous, or with relaxed tetragonality, characterized by low polarization and low capacitance [Hoshina *et al*, 2008].

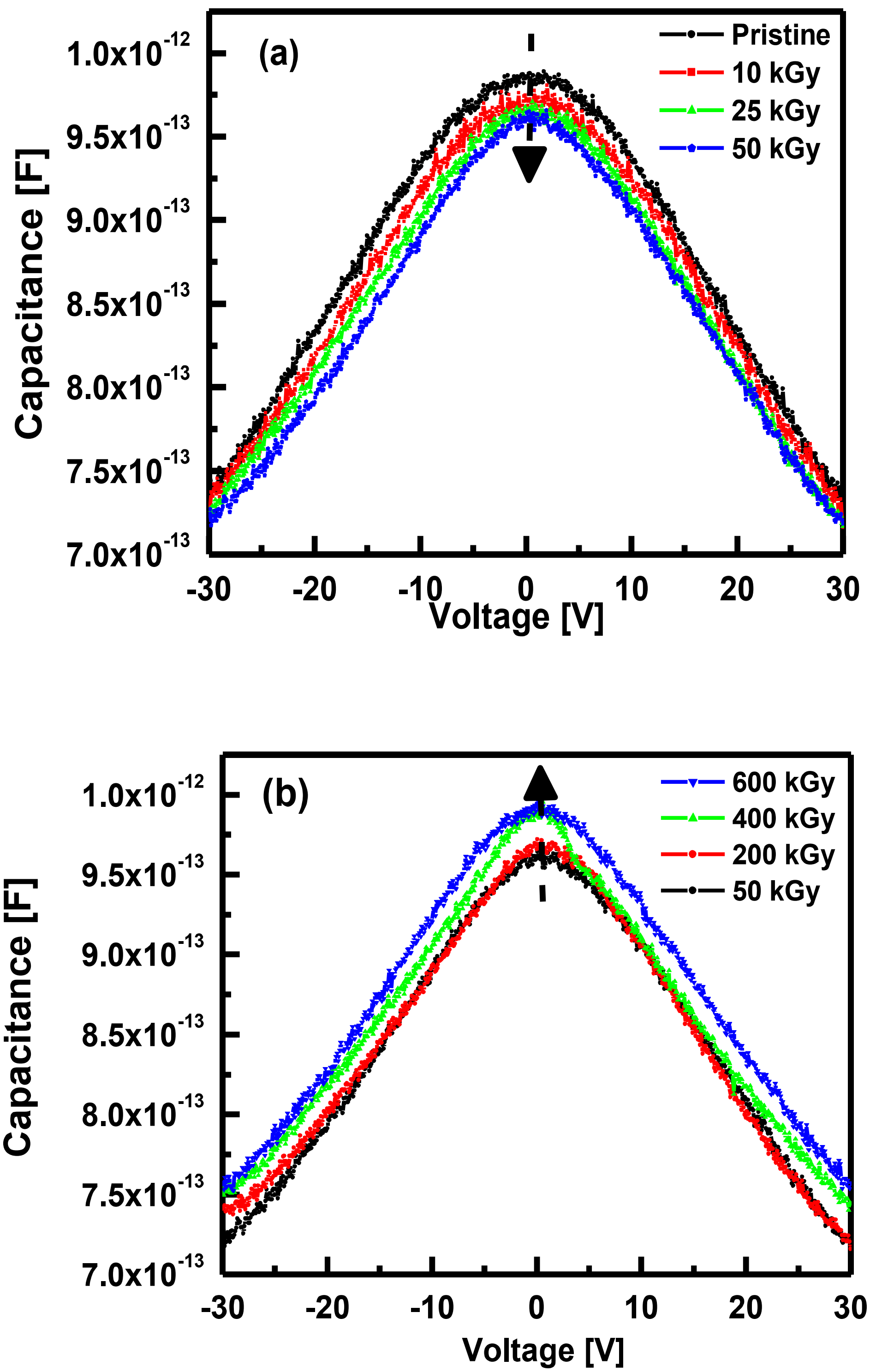

Figure 5.7: The capacitance as a function of the cumulative gamma doses from (a) 0 to 50kGy and (b) 50 to 600 kGy at 500 kHz frequency.

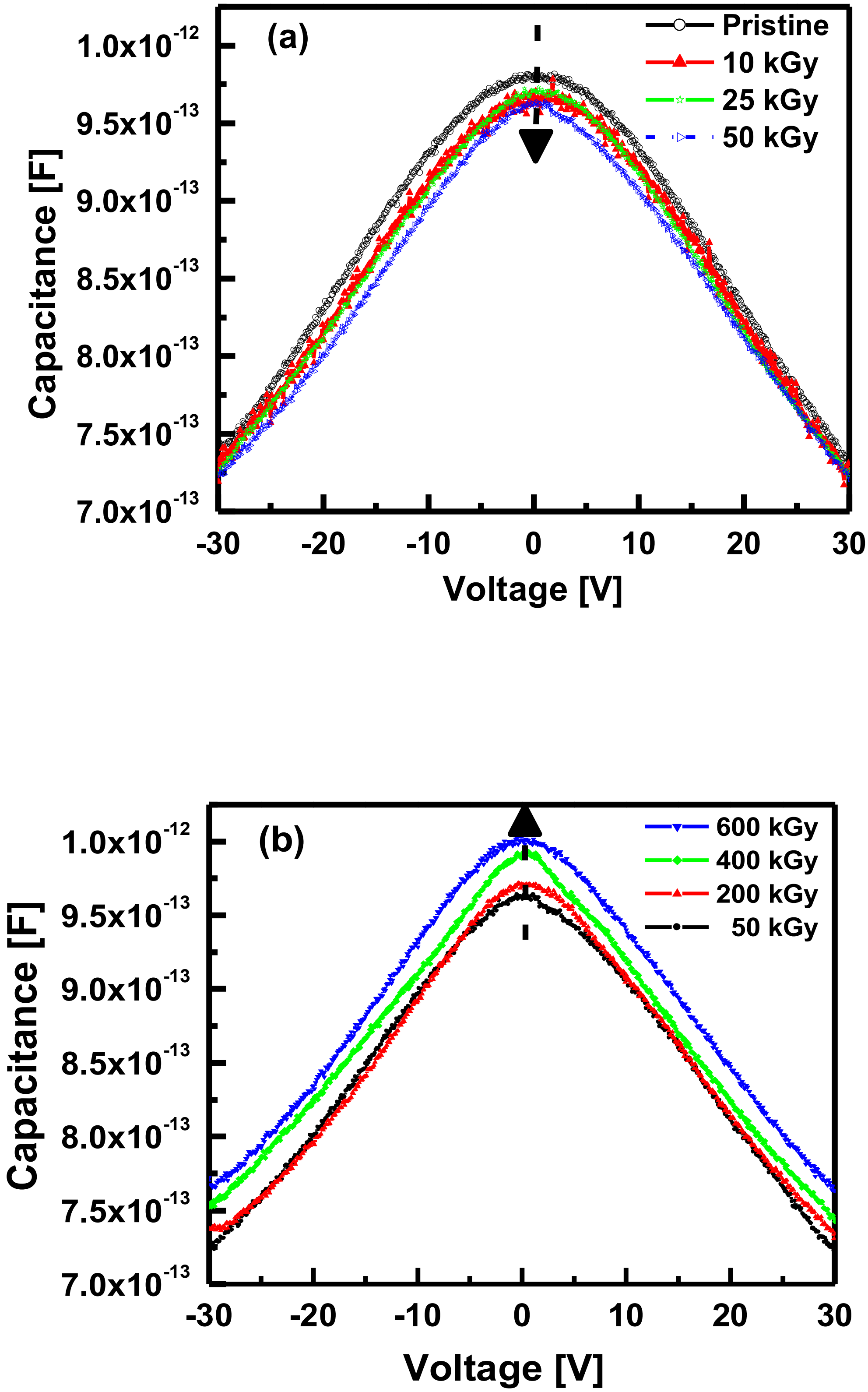

Figure 5.8: The capacitance as a function of the cumulative gamma doses from (a) 0 to 50kGy and (b) 50 to 600 kGy at 1 MHz frequency.

C-V measurements of the same IDC capacitor for higher cumulative doses (> 50 kGy) were also performed and shown in Figure 5.8. It was observed that the capacitance of the device is increased and reached higher than the virgin device for the cumulative dose of ≥400 kGy at the frequency of 1MHz, whereas capacitance at the frequency of 500 kHz was observed similar with the un-irradiated device at the peak value of capacitance. Conclusively the capacitance of

IDC device was found to decrease initially with increasing gamma dose and it starts to increase with gamma dose (≥100 kGy) to the maximum dose of about 600 kGy where the capacitance is more than the un-irradiated device. More importantly, the observed tunability is virtually the same as no significant difference was measured for pre and post irradiated device.

These results could be explained in combination of grain size effect, crystallinity, oxygen defects and mobile carriers generated during high radiation exposure. The mechanism of polarization process plays important role to increase the capacitance of the device after a certain gamma dose [Mane *et al*, 2011]. A gradual formation of mobile charge carriers or space charge polarisation where the electron exchange interaction may result in local displacement of the electron in the direction of electric field which in turn increases the capacitance at higher radiation exposure [Tataroglu and Altindal, 2007; Sinha *et al*, 2001; Al-karmi, 2006]. The well-known bell shaped structure is not distorted with the cumulative gamma dose upto 600 kGy, reflects high resistance of the BST based device against ionizing radiations. The observed changes may also be attributed with structural modification after gamma irradiation to the BST thin film. The changes in crystal structure along with space polarization after a certain gamma dose may be responsible for the increment in the capacitance.

5.4.2 Current-Voltage (I-V) Measurements

Analysis of dc leakage in thin films is of prime importance both in terms of fundamental and application points of view. The leakage current conduction in the ferroelectric thin films plays a major role in deciding device performance and the dielectric breakdown of the dielectric thin films. These parameters limit the functionality of the device, with respect to the applied field, gamma irradiation and dimension of the thin film. The presence of defects, grain boundaries and electrostatic potential barriers at the film-electrode interfaces dominate over the actual bulk current conduction phenomena in thin films. The leakage current analysis has always been essential requirement for tunable device applications to ensure that the flow of charge through the dielectric material remains negligible. The performance of the data storage device also depends on the ability of the active material to retain the stored data, which stores the information in terms of the charge [Scott *et al,* 1998.]. The various basic issues such as polarization switching, fatigue and imprint have already been addressed for several ferroelectric materials before integrating to the circuits. Another very important requirement for the consistency and quality is to retain low leakage current devices in the active material for the ferroelectric based devices. It has been found that high leakage current is accountable for the dielectric degradation of ferroelectric devices [Alemany *et al*, 1999]. In addition to the electrical conductivity measurements, charge movement is also correlated to the other dielectric measurements. Further in case of thin films, a very nominal voltage applied across the sample will produce the enormous amount of electric field owing to the small thickness associated with thin films.

I-V characteristics of BST thin films were studied on IDC structured device to investigate changes, before and after gamma irradiation at different total gamma doses. Conduction mechanisms in perovskite-type titanate thin films have previously been studied including $(Ba,Sr)TiO_3$ and $Pb(Zr,Ti)O_3$. It was concluded that the leakage current in Ssuch capacitor is controlled by the interfaces with the electrodes and the bulk of the film and one of them is highly dominant in most of the cases. Tunnelling and Schottky regimes govern the interface-controlled mechanisms and bulk-controlled mechanisms are governed by Poole Frenkel and space charge currents. Oxygen vacancies are generated inherently during the deposition of thin oxide film at the top BST electrode as well as in the BST film even without irradiation of the device. A small leakage current was observed in un-irradiated device, confirms the presence of bulk defects and interface states in the un-irradiated device and was in agreement with voltage and frequency dependence of un-irradiated IDC capacitor. Increases in leakage current resulted in dielectric loss as the dielectric loss is coupled with the resistive losses of the film [Cole *et al*, 2003]. There is enough experimental evidence to suggest that the dominant intrinsic defects in high-k oxide materials are related to oxygen related defect mechanism [McIntyre, 2007]. These intrinsic oxygen related defects are mainly believed to be present in the form of oxygen vacancies, oxygen interstitials, charged oxygen vacancies and oxygen deficiency.

Figure 5.9 shows I-V characteristics of the device for different cumulative doses and found that leakage current increases as a function of gamma dose. Gamma irradiation, results in the high density of acceptor like electron traps, preexisting traps and donor like traps including preexisting traps and generated traps in high-k dielectrics and has also been considered for changes in current voltage characteristics of the device [Zhao *et al*, 2006; Zhang *et al*, 2008]. Since oxygen vacancies and oxygen ions are generated while irradiation of the oxide film by gamma radiation. Similar phenomena has been reported for MIM high-k capacitor by Miao *et al* [Miao *et al*, 2009], considered impact of the generated vacancies on I-V characteristics. AFM data revealed decreasing in grain size for gamma irradiated device and consequently increases the grain density in turn reduces the space between grains which serves path to contribute in higher leakage current.

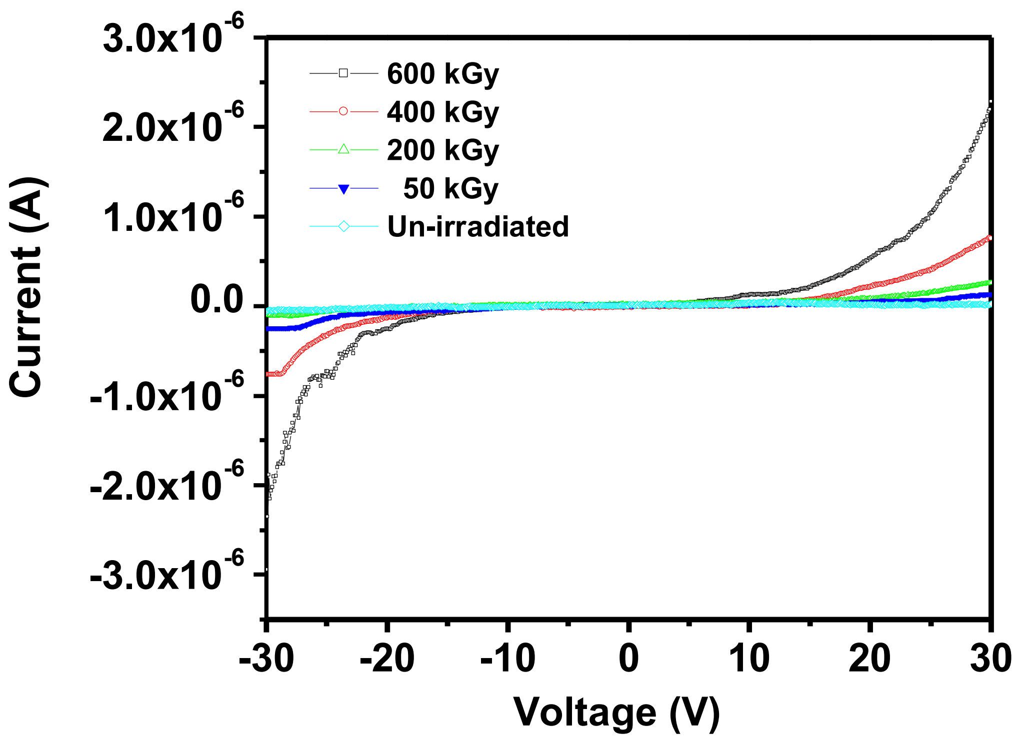

Figure. 5.9: Leakage current characteristics of IDC capacitor as a function gamma doses.

Gamma irradiation produces increased trap density, which give rise to changes in electric current of device. Enhanced leakage current can be attributed by increasing the trap density in the dielectric and charge carriers can hop between proximate traps leading to provide conducting paths through the dielectric film. Lowering schottky barrier height may be attributed by the oxygen vacancies (defect sites) at the surface of the film, which pins the Fermi level of the BST and metal (Au) electrode close to the conduction band minimum [Qin *et* al, 2003; Schafranek *et al*, 2010]. Possible explanation for the gamma irradiation dependence of the increasing current characteristics also includes forced diffusion of ionic species, results in charge transport phenomenon. In response to the applied field, these ions can move along grain boundaries or inside the grains. Changes in the electronic structure between grains including nano-regions of non-stoichiometry have also been proposed by Greuter *et al* [Greuter *et al*, 1990]. Charge state of ions, resulted by gamma irradiation may cause changes in the electronic characteristics therefore the interaction between the charge of the moving carrier and the non-periodic potential field will be altered at the interface between grains.

5.5 CONCLUSIONS

In this chapter, we have studied the effect of ^{60}Co gamma ray irradiation structural, morphological and electrical properties of the BST based tunable capacitors. The observed decrease in capacitance for low doses (≤ 50 kGy) and a noticeable increment above 50 kGy doses, indicates the presence of the different type of defects for different cumulative dose. Also increasing leakage current characteristics confirms the current serving path provided by grain boundaries along with increased number of radiation induced defects. The changes in the electrical properties may be attributed by the displacement damage introduced by irradiation to the device, causes to structural changes which have been confirmed from XRD and AFM. The increase in capacitance may be attributed to the presence of space charge polarization caused due to structural changes by high gamma irradiation. The XRD data shows diminishing and reproducing of the BST (110) peak with different gamma irradiation doses, which is correlated with the changes in capacitance value. AFM results confirm that grain size decreases with increasing gamma dose and attributed for higher leakage current of the device. The well-known bell shaped C-V characteristics did not distorted upto 600 kGy which suggest possible application of BST based tunable devices for radiation environment. The observed tunability (~25%) of the un-irradiated device was found nearly constant with gamma irradiation doses. This study showed that gamma ray induced defects play important role to the electrical properties of the devices and the BST based devices are highly resistant to gamma radiation, which reflects possible use in space and nuclear applications.