SYNTHESIS AND CHARACTERIZATION OF STRONTIUM STANNATE TITANATE THIN FILMS

May Kalayar Kyaing, Toe Toe Wai, Saw Moh Moh Oo and Moht Moht Than

Abstract
Strontium stannate titanate, Sr(Sn$_x$Ti$_{1-x}$)O$_3$ thin films on silicon substrates were successfully prepared by spin coating technique. The crystallographic properties of the films were analyzed by X-ray diffractometer and a cubic perovskite structure was found in all samples. The surface morphologies of the films were investigated using scanning electron microscope (SEM) and it was observed that fine grains with different sizes distributed the whole surface of the films. The dielectric constants as a function of applied electric field were calculated from capacitance-voltage measurements at the frequency range of 1 kHz to 100 kHz using LCR meter and the transition behavior was studied depend upon tin content.

Keywords: Sr(Sn$_x$Ti$_{1-x}$)O$_3$ thin films, SEM, XRD, dielectric constants

Introduction
Strontium titanate (SrTiO$_3$) with a cubic perovskite structure has been investigated intensively due to its unique dielectric and ferroelectric properties which are of great interest in the technological applications such as capacitors, actuators, and nonvolatile random-access memory devices. Strontium titanate (ST) has been of particular importance from the perspective of fundamental solid state physics, as well as in technological applications. Because of its good dielectric properties, it has become very usable in devices in integrated microelectronics and in memory storage. Also, the SrTiO$_3$ has gained additional practical importance as the most widely used substrate for depositing high–Tc superconducting films. As experiments on semitransparent samples are becoming more common, it is important to know the substrate dielectric function in order to correctly analyze the optical measurements performed on film plus the substrate system.

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Doping is often used to change the dielectric properties of the strontium titanate that arise as a result of the incorporation of different cations is essential. It has been well documented that a small number of impurity ions can change the properties of this interesting system. The variation in the lattice parameters of tin doped strontium titanate thin films with change in dopant concentration may be attributed to the change in ionic radius. The ionic radii of Sr$^{2+}$, Ti$^{4+}$ and Sn$^{4+}$ are 1.12, 0.68 and 0.69 Å respectively. Hence the ionic radius of Sn$^{4+}$ is approximately equal to that of Ti$^{4+}$ and hence tin can be easily diffused into the lattice and may substitute on B site atom in ABO$_3$ perovskite structure. The increased value of lattice parameters indicates that tin ion substitutes the B site of ABO$_3$ structure.

In the present research, the influence of doping by the Sn ions on the dielectric properties of the SST thin films were examined by a frequency range of 1kHz to 100kHz. The fabrication of high quality SST thin films with a high dielectric constant and low loss tangent is very important technological issue for practical frequency agile device and optical waveguide applications. In particular, non-linear dielectric and optical properties are suitable for application in the area of dynamic random access memory (DRAM) and optical waveguide applications. They are also used for frequency agile devices such as radio frequency (RF) switches, phase shifters, filters, delay lines, antennas and tunable microwave device applications.

**Experimental Details**

Tin doped strontium titanate, Sr(Sn$_x$Ti$_{1-x}$)O$_3$ thin films coated over silicon substrates with various dopant concentrations of ($x=0.1$ to 0.3) were prepared by sol-gel method. Coating of solution onto the substrates was performed by spin coating at 3000 rpm for 30 sec. Coated layers were dried in order to remove the solvent. The three different films were annealed at 700°C for 1 hr in a furnace to obtain crystallization. Scanning electron microscopy (SEM) were used to examine the surface grain morphologies of the strontium stannate titanate (SST) thin films. The phase and crystallinity of the films were characterized by X-ray diffraction (XRD) analysis with Cu-K$_{\alpha 1}$ radiation. The dielectric constants as a function of applied electric field were
calculated from capacitance-voltage measurements at the frequency range of 1 kHz to 100 kHz using LCR meter.

**Results and Discussion**

**X-ray diffraction (XRD) analysis**

X-ray diffraction pattern of tin doped strontium titanate thin films coated onto silicon substrates with various dopant concentrations are shown in Fig 1 (a-c). The characteristics peaks in the XRD patterns confirmed the presence of SST material and also indicated that all the films are well crystallized and cubic perovskite structure. The lattice parameters were slightly increased with the increasing of tin content. The variation in the lattice parameters of tin doped ST thin films with change in dopant concentration may be attributed to the change in ionic radius. The XRD patterns showed that the increased dopant concentration leads to decrease in intensity of diffraction peaks with preferred orientation at (110) planes. The crystallite size was calculated using Scherrer’s formula,

\[ D = \frac{0.9\lambda}{\beta \cos \theta} \]

The peak positions (2θ), intensity, full width half maximum, lattice parameters and crystallite sizes of all the films are listed in Table 1.

**Morphology and Compositional analysis**

The surface morphologies of the tin doped strontium titanate thin films were evaluated using SEM as shown in Fig 2 (a-c). These results showed a well-developed grain size and dense microstructure in all samples. Grain sizes of the samples are 0.45 μm, 0.67μm and 0.825 μm respectively. The average grain size increases with the increasing of tin content. The effect of doping on grain size is usually interpreted in terms of dopant solubility and distribution of doping ions between the surface and interior parts of the grain.
Dielectric Properties

The dielectric constants of strontium stannate titanate thin films were calculated from capacitance-voltage measurements at the frequency range of 1 kHz to 100 kHz. The dielectric constant varies with the applied voltage. The dielectric constant and dielectric loss of the films as function of applied voltage, Sn content and frequency are shown in Fig 3 (a-d). The maximum value of dielectric constant was occurred at the 10% tin doped strontium titanate thin film measured in a frequency range of 1 kHz. The dielectric constants of the films decrease with the increasing of dopant concentration and the results are listed in Table 2.

**Figure 1:** (a) X-ray diffraction of 10% tin doped ST thin film

**Figure 1:** (b) X-ray diffraction of 20% tin doped ST thin film
Figure 1: (c) X-ray diffraction of 30% tin doped ST thin film

Figure 2: (a) SEM image of 10% tin doped ST thin film
Figure 2: (b) SEM image of 20% tin doped ST thin film

Figure 2: (c) SEM image of 30% tin doped ST thin film
Figure 3: (a) The dielectric constant of three SST thin films as a function of tin content

Figure 3: (b) The dielectric constant of three SST thin films as a function of frequency
Figure 3: (c) The dielectric constant of three SST thin films as a function of applied voltage

Figure 3: (d) The dielectric loss of three SST thin films as a function of applied voltage
Table 1: The peak positions \( (2\theta) \), intensity, full width half maximum (FWHM), lattice parameters \( (a) \) and crystallite sizes \( (D) \) of SST thin films

<table>
<thead>
<tr>
<th>Thin Films</th>
<th>Peak positions ( (2\theta) )</th>
<th>Intensity (cps)</th>
<th>FWHM</th>
<th>a (nm)</th>
<th>D(nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10% tin doped ST</td>
<td>32.135</td>
<td>724</td>
<td>0.368</td>
<td>3.935</td>
<td>22.467</td>
</tr>
<tr>
<td>20% tin doped ST</td>
<td>32.139</td>
<td>662</td>
<td>0.412</td>
<td>3.936</td>
<td>20.068</td>
</tr>
<tr>
<td>30% tin doped ST</td>
<td>32.074</td>
<td>463</td>
<td>0.392</td>
<td>3.943</td>
<td>21.088</td>
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</tbody>
</table>

Table 2: The values dielectric constant and dielectric loss of SST thin films

<table>
<thead>
<tr>
<th>Thin Films</th>
<th>Dielectric constant</th>
<th>Dielectric loss (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10% tin doped ST</td>
<td>818.1279</td>
<td>0.4403</td>
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<tr>
<td>20% tin doped ST</td>
<td>689.1494</td>
<td>0.7891</td>
</tr>
<tr>
<td>30% tin doped ST</td>
<td>335.4931</td>
<td>0.9839</td>
</tr>
</tbody>
</table>

Conclusion

Strontium stannate titanate, SST thin films coated on silicon substrates with various dopant concentrations were prepared by sol-gel method. XRD patterns indicated that all the films are well crystallized and cubic perovskite structure. The lattice parameters were slightly increased with the increasing of tin content. SEM micrographs confirmed the crack free and uniform surface of the film. Grain sizes of the samples are in the range from 0.45\( \mu \)m to 0.825\( \mu \)m. The dielectric constant and dielectric loss of the films changed with different concentrations of tin. The maximum value of dielectric constant was occurred at the 10% tin doped strontium titanate thin film measured in a frequency range of 1 kHz. The polarization of the material is directly proportional to the dielectric constant of the material. Therefore, it is believed that tin doped strontium titanate thin films having high dielectric properties are good candidate materials for memory device applications.
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