Enhancement of breakdown strength of SrTiO₃/nano-SiO₂ composite film prepared by sol-gel technology

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Abstract: Dielectric (1-x)SrTiO₃/xSiO₂ (x=0, 0.01, 0.05 and 0.1) composite thin films were deposited onto Pt (100)/Ti/SiO₂/Si substrates via sol-gel and spin coating technology and their surface morphology was examined by field emission scanning electron microscopy, indicating that surface morphology was apparently influenced by the nano-SiO₂ addition. A broad adsorption band at 3200-3500 cm⁻¹ attributed to stretching vibrations of O-H bonds in the absorbed water was observed in fourier transform infrared spectroscopy (FTIR). The X-ray photoelectron spectroscopy (XPS) results show that nano-SiO₂ particles facilitate water absorption of STO thin films. The breakdown strength of SrTiO₃/nano-SiO₂ composite films is increased to about 332 MV/m. At the same time, the leakage current of 0.9SrTiO₃/0.1SiO₂ composite thin films decreases by two orders of magnitude under high electric field. Above results show that nano-SiO₂ particles facilitate water absorption of the thin films, making SrTiO₃ act as an effective solid state electrolyte for the anodic oxidation to achieve higher breakdown strength.

Keywords: SrTiO₃/nano-SiO₂; Composite film; Absorbed water; Anodic oxidation; Breakdown strength; Sol-gel technology

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1. Introduction

In recent years, dielectric thin films have received great attentions and become one of the predominant materials in electronic industry. SrTiO_3 (STO) based thin film is a promising candidate for many electric applications owing to its high relative permittivity, low loss, stability with various technologically relevant substrates and optical transparency [1-3]. Therefore, these excellent dielectric properties make STO a candidate for high energy storage dielectric material employed in solid state capacitors [4, 5].

It is well known that high energy storage density is dependent on high relative permittivity and breakdown strength [6]. In the past decades, many investigations have been carried out to improve the energy storage density by enhancing the relative permittivity and breakdown strength. Wang, Z. et al. [3] found that 0.4 mol% ZrO_2 addition improved the breakdown strength and energy density of SrTiO_3 ceramics due to the decreased grain size and homogeneous distribution. Chao, S. et al. [7] studied BaTiO_3 - SrTiO_3 layered dielectrics and revealed that layered structures offered flexibility to maximize the energy storage capability by tailoring the dielectric properties through varying the spatial configurations of BaTiO_3 and SrTiO_3 films. For linear dielectrics, energy storage density is proportional to the square of breakdown strength. Therefore, high breakdown strength makes a more pronounced contribution to the energy storage density. Moreover, the breakdown strength and leakage current had been observed to have a strong dependence on the electrode materials [8, 9]. Anodic oxidation reaction was found in the sol-gel thin films with valve metal electrodes, especially the anodization of Al electrodes [10, 11]. Under the driving

force of the applied electric field during the anodic oxidation process, metal cations and $\text{OH}^-/\text{O}^{2-}$ anions migrate towards the opposite directions, resulting in growth of the anodized oxide layer at the metal/dielectric interface. Hence, the formation of the anodized oxide significantly enhances the breakdown strength of dielectrics. Besides, it is worth noting that $\text{OH}^-/\text{O}^{2-}$ anions are major contributors to the anodic oxidation.

Silica is widely studied by many researchers for applications in varieties of fields. Ben Salem, M, K. et al. [12] found that nano- SiO_2 particles had an impact on the enhancement of the superconducting properties of $\text{YBa}_2\text{Cu}_3\text{O}_y$. In lithium-ion batteries, nano- SiO_2 embedded SiO_x matrix exhibited a stable discharge capacity with excellent cyclic stability and rate capability [13]. Moreover, silica additives led to an obvious improvement in the breakdown performance of SrTiO_3 ceramics [14]. Nano- SiO_2 particles are of extremely high surface activity and porous structure [15]. There are a large number of unsaturated bonds and hydroxyl groups on the surface of nano- SiO_2 particles, resulting in sufficient water absorbing capacity [15-17]. Typically, silica was used as one of basic materials for humidity sensors [18-20]. Basing on the above, it is interesting to see if nano- SiO_2 particles can be introduced into STO thin films to enhance the breakdown strength by improving the efficiency of the anodic oxidation through providing sufficient $\text{OH}^-/\text{O}^{2-}$ anions.

2. Material and methods

$(1-x)\text{SrTiO}_3/x\text{SiO}_2$ ($x=0, 0.01, 0.05$ and 0.1) composite films (abbreviated as ST/S0, ST/S1, ST/S5, ST/S10, respectively) were fabricated by sol-gel and spin-coating technology. For preparation, semi-hydrate strontium acetate ($\text{Sr}(\text{CH}_3\text{COO})_2 \cdot 0.5\text{H}_2\text{O}$) and titanium

isopropoxide ($\text{Ti}(\text{OCH}(\text{CH}_3)_2)_4$) were used as strontium precursor and titanium precursor, respectively. SiO_2 particles (average size ~ 20 nm) dispersed in ethylene glycol was used as dopant. Firstly, strontium acetate was dissolved in acetic acid with constant stirring for 30 min at 80°C . Next, polyvinyl pyrrolidone (PVP) acetic acid solution was added at 50°C , forming solution A. At the same time, titanium isopropoxide was dissolved in ethylene glycol monomethyl ether with continuously stirring at 50°C . Then, acetylacetone acting as chelating agent was added to restrain the hydrolysis of titanium isopropoxide, forming solution B. Subsequently, solution B was added in drop-wise to solution A under constant stirring. Then, ethylene glycol was added to adjust the viscosity and prevent the films from cracking. Afterwards, colloidal dispersion of SiO_2 nanoparticles was added into the mixture. At last, the obtained solution was maintained under constant stirring for 3 h and then aged for 3 days.

Thin films were deposited on Pt (100)/Ti/ SiO_2 /Si substrates by spin-coating with a rotating speed of 3000 rpm for 20 s. After each deposition, the wet films were dried in a tubular furnace at 200, 350, 500°C for 5 min successively to form solid films by evaporating the solvent and burning out the organic residuals. Then, samples with 7 layers were annealed at 500°C for 3 h in a tubular furnace in flowing oxygen atmosphere. Finally, Al top electrodes with the thickness of 90 nm and 160 nm, and diameter of 1 mm were fabricated by magnetron sputtering for electrical measurements.

The surface morphology of the films was observed by field emission scanning electron microscopy (FESEM, S-4700, HITACHI, Japan), and the thickness was measured by the

cross-sectional FESEM images. The roughness of the thin films was investigated by an atomic force microscope (AFM, SPA 400, SPI3800N, Seiko, Japan). The chemical composition of the films was observed by fourier transform infrared spectroscopy (FTIR, EQUINOX 55, Bruker Optics, Germany). The chemical states of silicon and oxygen in the thin films were analyzed by X-ray photoelectron spectrometer (XPS, ESCALAB 250Xi, Thermo Fisher, America). The current as a function of the electric field was measured using a Keithley 2400 source meter unit interfaced with a computer to record data. Positive voltage in a successive voltage step of 0.2 V with delay time of 0.1 s was applied to Al top electrodes until the ultimate breakdown happened. *I-E* characteristics were measured at a temperature of 25 °C and relative humidity of 30% in a humidity chamber (GP/TH-50, Guangpin, China). The morphology of the samples after electrical measurements was examined by a metallographic microscope (BX51, OLYMPUS, Japan).

3. Results and discussion

Fig. 1 shows surface morphology of $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ ($x=0, 0.01, 0.05$ and 0.1) composite films. It can be seen that the surface of the ST/S0 thin film is highly smooth and homogeneous. However, the surface morphology obviously changes with the increase of the amount of nano-SiO₂ particles, especially for the ST/S10 composite thin film. AFM was employed to further investigate the surface morphology of $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ films, as shown in Fig. 2. A large number of fine particles are observed in $\text{SrTiO}_3/\text{nano-SiO}_2$ composite thin films. The surface roughness is analyzed to be 0.7, 2.4, 4.0 and 4.5 nm, respectively. The toughness increases with the increase of the amount of nano-SiO₂ particles,

which is in accordance with the results reported by Yang, H. et al [21]. As a consequence, the surface morphology is significantly influenced by the nano-SiO₂ addition. And the thickness are 303, 286, 276, 299 nm for the ST/S0, ST/S1, ST/S5 and ST/S10 films, respectively.

Fig. 3a shows the typical *I-E* characteristics of (1-x)SrTiO₃/xSiO₂ composite thin films with 160 nm thick Al top electrodes. At low electric field region (0-110 MV/m), the leakage current slightly increases with the addition of nano-SiO₂. However, the leakage current significantly decreases under high electric field. Typically, the leakage current of the ST/S10 composite film is approximately two orders of magnitude lower than that of the ST/S0 thin film. In addition, the breakdown strength is enhanced by adding nano-SiO₂ particles. In order to figure out the effect of the nano-SiO₂ particles on breakdown characteristics of STO thin films, weibull distribution of the breakdown strength is illustrated in Fig. 3b. It can be seen obviously that the breakdown strength is significantly improved with the increasing nano-SiO₂ content. Especially, the breakdown strength of the ST/S10 composite film is increased by 33% up to 332 MV/m compared with the ST/S0 film.

Structural characteristics of (1-x)SrTiO₃/xSiO₂ composite films were investigated to explain the above phenomena. Fig. 4 shows the FT-IR spectra of (1-x)SrTiO₃/xSiO₂ composite thin films. The peaks at 640 cm⁻¹ and 1112 cm⁻¹ are corresponding to the stretching vibration modes of Ti-O and Si-O-Si bonds, respectively [22-24]. The peak at 1450 cm⁻¹ can be assigned to the chemisorbed CO₂ molecules, while the peak at 2350 cm⁻¹ is closely related to the physisorbed CO₂ molecules [25]. The peak at 2920 cm⁻¹ is attributed

to C-H bonds [26]. It is highly notable that a broad absorption band at 3200-3500 cm^{-1} is observed, and it is attributed to the stretching vibrations of O-H bonds in the absorbed water [27], which provides OH/O^{2-} anions for the anodic oxidation reaction at the interfaces between $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ films and Al electrodes [10].

XPS, as a powerful technology to investigate the chemical composition of the thin films, was also employed to explore the chemical bonding of silicon. Fig. 5 displays the XPS spectrum of Si2p. It can be observed that Si2p has two peak positions, suggesting that two silicon species present on the surface. The peaks at the binding energy of 103.6 eV and 101.9 eV are attributed to SiO_2 and SiO_x , respectively [28-30]. It can be concluded that silicon exists not only in the form of SiO_2 , but also in the form of SiO_x , which is in accordance with the previous reports [15-17] that there are a large number of unsaturated bonds and hydroxyl groups on the surface of nano- SiO_2 particles.

The O1s high-resolution XPS spectra of the ST/S0 and ST/S10 thin films were further analyzed to investigate the influence of the nano- SiO_2 particles on chemical composition of STO thin films. There are three oxygen species on the surface of the thin films according to the spectra of O1s in Fig. 6. The peak at 530.3 eV is attributed to O^{2-} ions in the crystalline structure of STO [31]. The peak at 533.4 eV is usually linked to the loosely bound oxygen on STO surface [32]. It should be noted that the middle peak at the binding energy of 531.8 eV is closely related to the hydroxyl groups in the absorbed water of STO films [33]. The XPS results strongly confirm the existence of the absorbed water in the sol-gel thin films, which is in accordance with FTIR results. To explore the influence of the nano- SiO_2

particles on water absorbing capacity of STO films, the concentration ratio of hydroxide (hydroxide/total) in the films is estimated. Supposing the full width at half maximum (FWHM) of the three corresponding O1s peaks of the ST/S0 and ST/S10 films are the same. The hydroxide/total ratios of the ST/S0 film and the ST/S10 composite film are analyzed to be 17% and 29%, respectively. It suggests that the nano-SiO₂ particles facilitate water absorption of STO thin films, which is the main contributing factor to improve the efficiency of the anodic oxidation, resulting in enhanced electrical characteristics.

The 90 nm thick Al top electrodes were deposited to corroborate the effect of the nano-SiO₂ particles on the anodic oxidation. *I-E* plots of (1-x)SrTiO₃/xSiO₂ films were measured, as shown in Fig. 7. It is obvious that the breakdown strength increases with the increase of nano-SiO₂ content, except for the ST/S10 composite film. It is worth noting that the ultimate breakdown was not observed with the leakage current abruptly decreasing from 30 μ A to 2 nA at about 214 MV/m. To further investigate the above phenomena, the surface morphology of the samples after electrical measurements was observed using a metallographic microscope (Fig. 8). For ST/S0, ST/S1 and ST/S5 samples, only the edges of the Al electrodes become purple. However, for the ST/S10 sample, all the grayish surface area of the Al electrode has turned into purple, owing to the occurrence of the anodic oxidation [10]. With the progress of the anodic oxidation, an anodized aluminum oxide (AAO) layer starts to grow at the cost of the Al electrode. Once the Al electrode completely transforms into aluminum oxide, it is no longer conductive, leading to a sudden decrease of the leakage current. It can be deduced that the efficiency of the ST/S10 composite film is

much higher than the other samples.

As shown in Fig. 9a, there are a large number of various hydroxyl groups on the surface of nano-SiO₂ particles [15-17], contributing to the sufficient water absorbing capacity of the composite film. The absorbed water of the STO/nano-SiO₂ composite film is supposed to be the supplier of OH⁻ and O²⁻ anions to the anodic oxidation. The anodic oxidation model of the STO/nano-SiO₂ composite thin film is illustrated in Fig. 9b. Platinum, the bottom electrode, serves as the cathode of the STO/nano-SiO₂ composite film. And aluminum, the top electrode, serves as the anode. Under the driving force of the electric field, the OH/O²⁻ anions in the STO/nano-SiO₂ composite film migrate towards the aluminum anode, and the aluminum metal at the interface of Al/STO is transformed from Al⁰ atoms into Al³⁺ cations. OH/O²⁻ anions tend to combine with Al³⁺ cations forming aluminum oxide and/or hydroxide. With the progress of the anodic oxidation, an AAO layer grows at the interface of Al/STO. While nano-SiO₂ particles, possessing high water absorbing capacity, provide sufficient OH/O²⁻ anions for STO thin films and improve the efficiency of the anodic oxidation. Moreover, it is well known that the AAO film is a high quality dielectric material with the breakdown strength up to about 700-1000 MV/m [34, 35]. Therefore, nano-SiO₂ particles significantly improve the breakdown strength of STO films.

4. Conclusions

In summary, we have systematically investigated the effect of the nano-SiO₂ particles on the breakdown behaviors and leakage current of amorphous STO thin films fabricated by sol-gel technology. The breakdown characteristics and leakage current are significantly

enhanced by the nano-SiO₂ addition. Particularly, for the ST/S10 composite film, the breakdown strength is increased by 33%, moreover the leakage current is about two orders of magnitude lower under high electric field compared with the ST/S0 thin film. The excellent electrical properties are attributed to the improvement of anodic oxidation efficiency. Nano-SiO₂ particles, with a large number of unsaturated bonds and hydroxyl groups on the surface, facilitate water absorption of the thin films, making STO act as an effective solid state electrolyte for the anodic oxidation. Based on the above results, it can be concluded that the nano-SiO₂ addition may be a promising way to enhance electrical properties of dielectric films.

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Figure captions:

Fig.1 FESEM micrographs of $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ composite films.

Fig.2 AFM images of $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ composite films: (a) ST/S0; (b) ST/S1; (c) ST/S5 and (d) ST/S10.

Fig.3 (a) I - E Plots for $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ composite films with 160 nm thick Al top electrodes; (b) Weibull distribution of the breakdown strength of $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ composite films. Insert shows the dependence of breakdown strength on nano-SiO₂ molar fraction of $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ composite films (i is the number of the specimen, n is the sum of specimens and E is the breakdown strength).

Fig.4 FTIR spectra of $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ composite films.

Fig.5 XPS spectrum of Si2p for ST/S10 composite film.

Fig.6 XPS spectra of O1s for: (a) ST/S0 thin film and (b) ST/S10 composite film.

Fig.7 I - E Plots for $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ composite films with 90 nm thick Al top electrodes.

Fig.8 Metallographic photographs of $(1-x)\text{SrTiO}_3/x\text{SiO}_2$ composite films with 90 nm thick Al top electrodes after electrical **measurements**.

Fig.9 (a) The surface characteristics of nano-SiO₂ and (b) The anodic oxidation reaction model of the $\text{SrTiO}_3/\text{nano-SiO}_2$ composite film with Al electrode under high electric field (S·H is $\text{SiO}_2 \cdot n\text{H}_2\text{O}$, AAO is anodic aluminum oxide).

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