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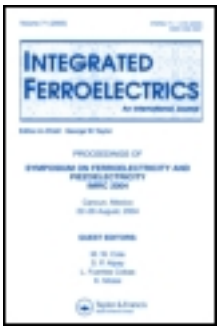

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Electrical properties of O_2 and N_2 annealed (Ba, Sr)TiO_3 thin films

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Electrical Properties of O_2 and N_2 Annealed (Ba,Sr)TiO_3 Thin Films

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The dielectric constant and the leakage current of (Ba,Sr)TiO_3 (BST) thin films deposited on Pt bottom electrodes before and after annealing in O_2 and N_2 ambients were investigated. The crystallinity was improved after post-deposition annealing. The refractive index, dielectric constant, and leakage current of the films were strongly dependent on the annealing conditions. The O_2-annealed BST films have higher dielectric constant and lower leakage current than those annealed in N_2 ambient.

Keywords: BST; annealing; oxygen vacancy; leakage current

INTRODUCTION

In recent years, thin films made from high dielectric constant materials have attracted great attention for use in fabricating cell capacitors for high density dynamic random access memories (DRAMs), since employment of high-dielectric constant films can lower the height of the storage node and simplify the cell structure. The (Ba,Sr)TiO_3 (BST) films have been reported to be the most promising capacitors materials for giga-bit DRAMs because of its high dielectric constant, low leakage current density, high dielectric breakdown strength, paraelectric perovskite phase that does not exhibit fatigue and aging and the ease of compositional control [1-4]. Pt is the most studied electrode material because it has a large work function which lowers the leakage current

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strength, paraelectric perovskite phase that does not exhibit fatigue and aging and the ease of compositional control [1-4]. Pt is the most studied electrode material because it has a large work function which lowers the leakage current of BST capacitors. In this paper we have studied the electrical characteristics of annealed BST films on Pt. Electrical and reliability characteristics of BST films annealed at different conditions are compared.

EXPERIMENTAL

Metal-BST-metal capacitors were used as test devices. Pt bottom electrode with thickness of 100 nm was directly deposited on SiO_2 coated Si-substrates by RF magnetron sputtering at 350 ℃. BST thin films (100 nm) were deposited by RF magnetron sputtering at 450 and 27 ℃ in ambient of 50% O_2 and 50% Ar. After BST deposition, some samples were treated in O_2 and N_2 at 550, 650 and 750℃ for 1 to 3 min using rapid thermal annealing and 30 min using furnace annealing. Finally, Pt (50 nm) top electrodes with diameters of 165, 255 and 350 μm were formed by electron beam evaporation through a shadow mask. The refractive index and the film thickness of BST films before and after annealing were measured by Rudolph Auto EL-III ellipsometer at a wavelength of 6328 Å.

RESULTS AND DISCUSSION

Fig. 1 depicts the change of refractive index with annealing time for BST thin films deposited at 27 ℃. The refractive index of BST annealed at 550 ℃ approaches a constant value ~ 1.85. The refractive indexes of BST annealed at 650 and 750℃ increase with increasing annealing time, but the index of BST annealed at 750 ℃ attains saturation when annealed over 1 min.

Fig. 2 shows the variation of thickness with annealing time for BST thin films deposited on Pt at 27℃. The film thickness of BST annealed at 550 and 650℃ decreases with increasing annealing time. Also the film thickness of BST annealed at 750℃ decreases with increasing annealing time, but it attains saturation when annealed over 1 min. This thickness reduction can be explained as the result of an increased density and elimination of porosity [5].

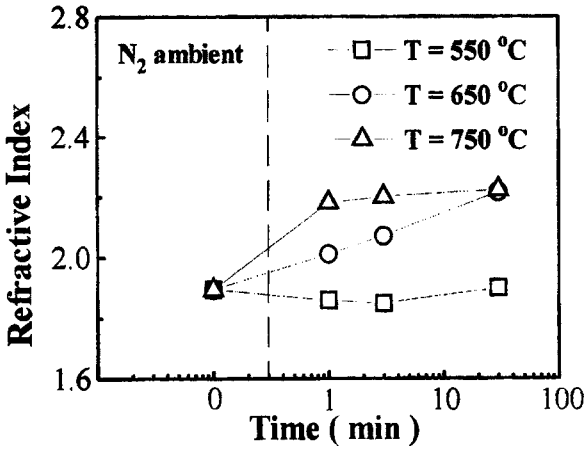

FIGURE 1 The refraction index of BST films deposited at 27 °C as a function of annealing time in N_2.

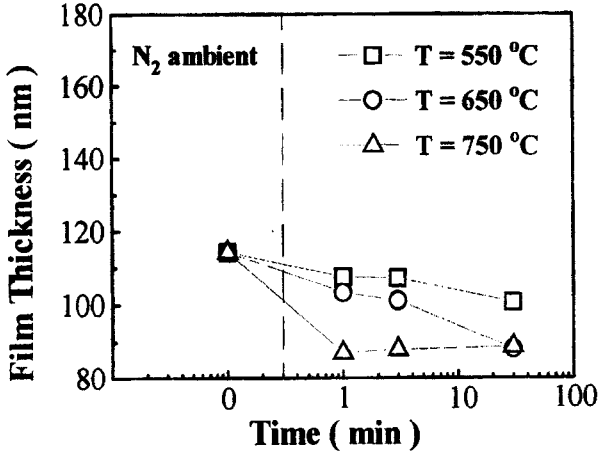

FIGURE 2 The thickness of BST films deposited at 27 °C as function of annealing time in N_2.

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Fig. 3 indicates that the dielectric constant of 27 ℃-deposited BST after annealed at 550 ℃ in N_2 ambient approachs a constant value ~ 23. The dielectric constant of BST annealed at 650 and 750 ℃ increases with increasing annealing time, which may be due to the increase of crystallinity with annealing. However, the dielectric constant of BST annealed at 750℃ attains saturation after annealing over 1 min. The results shown in Fig. 3 are in good agreement with those indicated in Fig. 1. Fig. 4 shows that the dielectric constant of BST annealed in O_2 have the similar result with Fig. 3. The dielectric constant of BST annealed in O_2 is larger than that of BST annealed in N_2. These results may be attributed to the lowering of the oxygen vacancy concentration in BST annealed in O_2, which may lead to an increase in dielectric constant.

Fig. 5 illustrates the leakage current measured at 100 kV/cm (with 30 sec delay time) as a function of N_2 annealing time. The leakage current of BST annealed at 550 ℃ decreases with increasing annealing time, because of increasing densification of BST films after post-annealing, as shown in Fig. 2. The leakage current of BST annealed at 650 ℃ decreases with increasing annealing time, but it slightly increases for BST films annealed for 30 min. The possible reason is that the increase in densification of BST films after post-annealing leads to a decrease of leakage current, but the increase of crystallinity for annealing over 30 min will increase the leakage current. The leakage current of BST annealed at 750℃ increases with increasing annealing time, because the crystallinity effect is more dominant, as compared to the effect of densification. Also, morphology changes such as hillocks can greatly influence leakage after annealing at high temperature or long time; or increase in oxygen-vacancy concentration as well. Fig. 6 shows that the leakage current of BST annealed in O_2 has the similar behavior as that in Fig. 5. The leakage current is slightly increased for BST annealed in O_2 at 650 ℃ over 3 min, whereas after annealed in N_2 at 650℃ for over 30 min (Fig. 5) the leakage current was also slightly increased. The dielectric constants of BST annealed in O_2 (Fig. 4) are larger than those of the BST annealed in N_2 (Fig. 3), and the leakage currents of BST annealed in O_2 (Fig. 6) are smaller than that in N_2

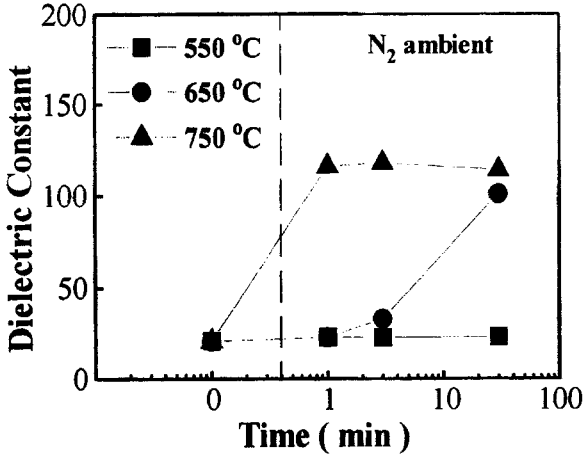

FIGURE 3 The dielectric constant of 27 °C -deposited films as a function of annealing time in N_2.

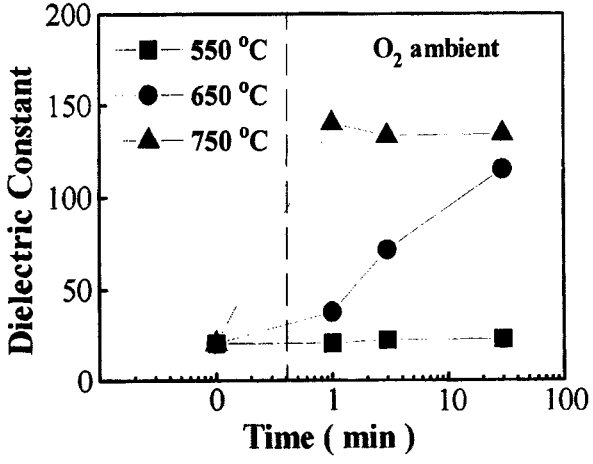

FIGURE 4 The dielectric constant of 27 °C -deposited films as a function of annealing time in O_2.

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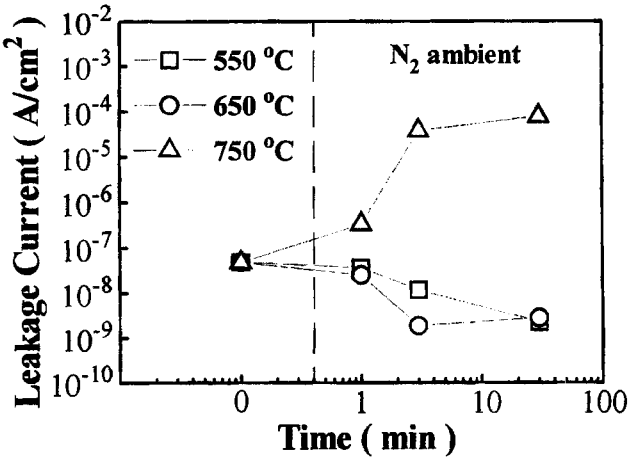

FIGURE 5 The leakage current of 27 °C -deposited films as a function of annealing time in N_2, at 100 kV/cm.

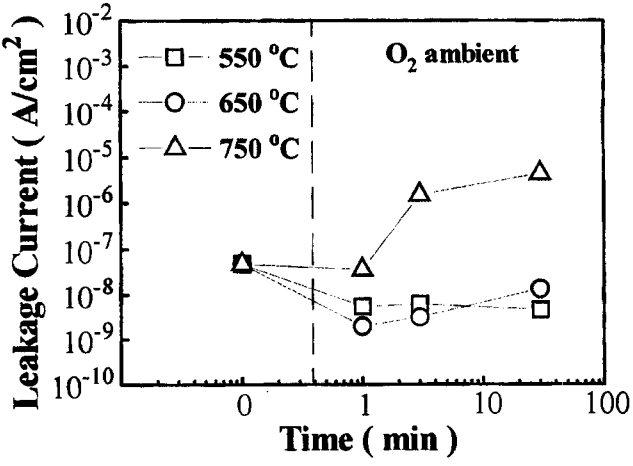

FIGURE 6 The leakage current of 27 °C -deposited films as a function of annealing time in O_2, at 100 kV/cm.

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(Fig. 5). The reason may be due to the decrease of oxygen vacancies of BST after annealed in O_2, and hence the dielectric constant can be enhanced and the leakage current can be suppressed. These results suggest that the BST films annealed in O_2 at 650 ℃ is a suitable process condition for obtaining high dielectric constant and low leakage current.

Fig. 7 shows that the dielectric constant of BST deposited at 27℃ and 450 ℃ and annealed at 650℃ in N_2 and O_2. The dielectric constant of BST films increases with increasing annealing time. The dielectric constant of 450 ℃ -deposited BST annealed at 650℃ in O_2 is larger than that of the BST annealed in N_2. Therefore, the polarization or crystallization of BST annealed in O_2 ambient is expected to be more enhanced than in N_2 ambient. Fig. 8 shows the leakage current of 27℃- and 450℃-deposited BST annealed at 650 ℃ in N_2 and O_2. The leakage current of O_2-annealed BST does not change with annealing time, probably due to the balance of crystallization effect and the reduction of oxygen-vacancy concentration. The leakage current of N_2-annealed BST increases slightly with increasing annealing time, because higher crystallization of N_2-annealed BST has enhanced the electrons transport and there is no compensation of reduction of oxygen vacancy concentration. The dielectric constant of BST films deposited at 450 ℃ after annealing in O_2 and N_2 ambient at 650 ℃ for 30 min are 476 and 390, respectively. The leakage current density of BST films deposited at 450 ℃ after annealing in O_2 and N_2 ambient at 650 ℃ for 30 min are 2.5×10^{-8} and 1.35×10^{-7} A/cm^2, respectively.

Fig. 9 shows the leakage current of as-deposited BST films (27℃ and 450 ℃). The leakage current at positive bias of BST deposited at 27℃ is larger than that of BST deposited at 450℃, because the BST film deposited at 27℃ is less dense than at 450 ℃. Fig. 10 depicts the leakage current of BST deposited at 450℃ and annealed at 650 ℃ in N_2 and O_2 for 3 min. The leakage currents in both conditions have almost the same value. Fig. 11 shows the leakage current of BST deposited at 450℃ under 650℃ annealing in N_2

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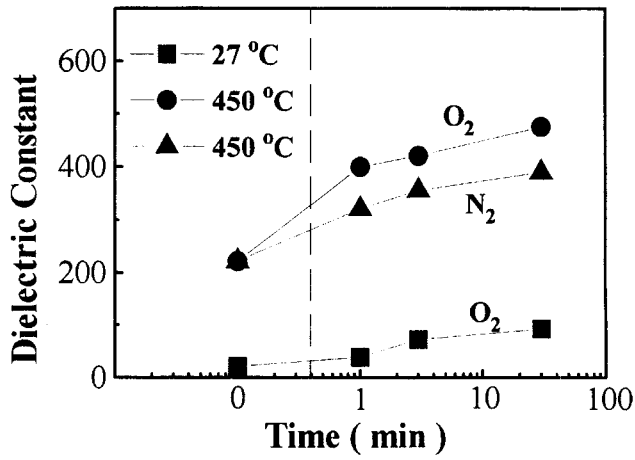

FIGURE 7 The dielectric constant of BST films deposited at 27 °C and 450 °C as a function of annealing time at 650 °C with O_2 and N_2 ambient as a parameter.

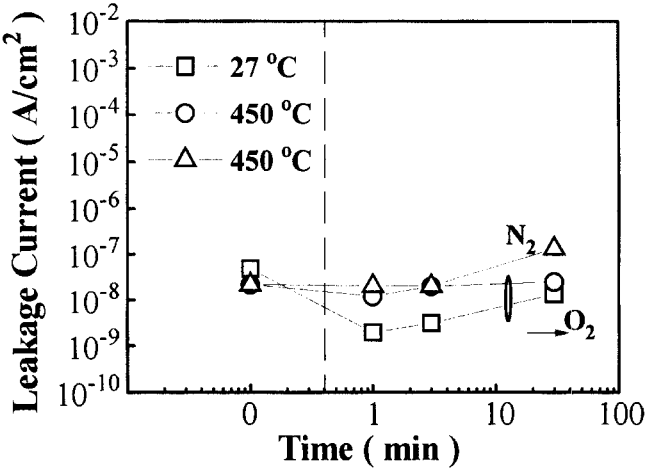

FIGURE 8 The leakage current of BST films deposited at 27 °C and 450 °C as a function of annealing time at 650 °C with O_2 and N_2 ambient as a parameter.

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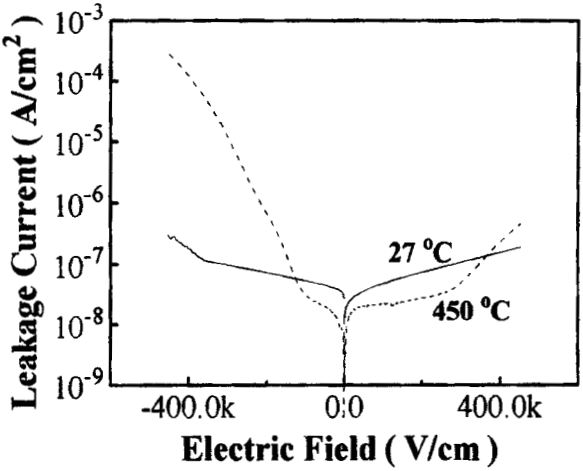

FIGURE 9 The leakage current of BST films deposited at 27 °C and 450 °C.

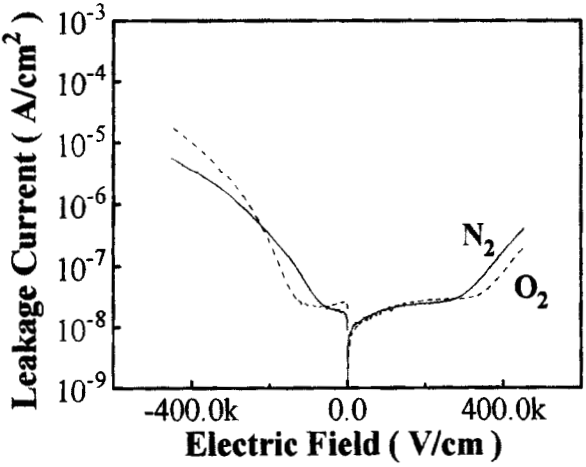

FIGURE 10 The leakage current of 450 °C-deposited BST films after annealed for 3 min at 650 °C in O_2 and N_2 ambient .

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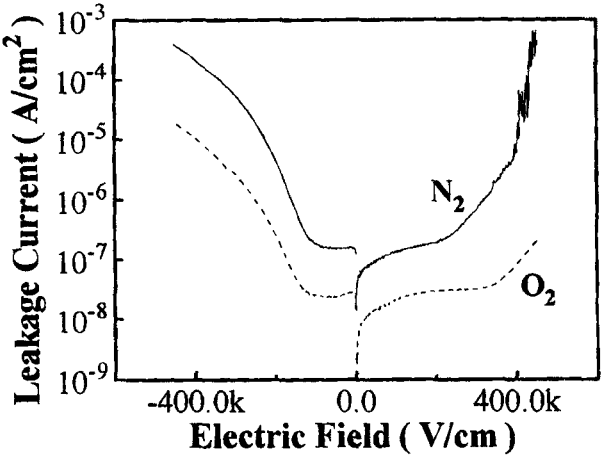

FIGURE 11 The leakage current of 450 °C-deposited BST films after annealed for 30 min at 650 °C in O_2 and N_2 ambient.

and O_2 for 30 min. The leakage current of O_2-annealed BST is smaller than that of N_2-annealed BST. Again, O_2 ambient is believed to reduce the oxygen vacancies, hence the leakage current of O_2-annealed BST is smaller than that of BST annealed in N_2.

We have also examined the BST films deposited on Pt bottom electrode at 27 °C and annealed in N_2O ambient. The capacitors showed a very large leakage current and therefore their dielectric constant could not be measured.

CONCLUSIONS

The dielectric constant and the leakage currnet of O_2- and N_2- annealed BST thin films were examined. The post-annealing process can improve the crystallinity of BST films. The dielectric constants of BST films deposited at 450 °C after annealing in O_2 and N_2 ambient at 650 °C for 30 min are 476 and 390, whereas the leakage currents of the films are 2.5×10^{-8} and 1.35×10^{-7} A/cm^2, respectively. The BST films annealed in O_2-ambient have a higher

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dielectric constant and a lower leakage current as compared to the films annealed in N_2-ambient. This may be attributed to the decrease of oxygen vacancies of O_2- annealed BST.

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