Conduction Mechanisms in Barium Tantalates Films and Modification of Interfacial Barrier Height

Yun-Hi Lee, Young-Sik Kim, Dong-Ho Kim , Byeong-Kwon Ju, *Member, IEEE*, and Myung-Hwan Oh, *Member, IEEE*

***Abstract*—The leakage current–voltage characteristics of rf-magnetron sputtered $BaTa_2O_6$ film in a capacitor with the top aluminum and the bottom indium-tin-oxide electrodes have been investigated as a function of applied field and temperature. In order to study the effect of the surface treatment on the electrical characteristics of as-deposited film we performed an oxygen plasma treatment on $BaTa_2O_6$ surface. The dc current–voltage, bipolar pulse charge-voltage, dc current-time, and small ac signal capacitance-frequency characteristics were measured to study the electrical and the dielectric properties of $BaTa_2O_6$ thin film. All of the $BaTa_2O_6$ films in this study exhibited a low leakage current, a high breakdown field strength (3–4.5 MV/cm), and a high dielectric constant (20–30). From the temperature dependence of the leakage current, we could conclude that the dominant conduction mechanism under high electrical fields (>1 MV/cm) is ascribed to the Schottky emission while the ohmic conduction is dominant at low electrical fields (<1 MV/cm). Furthermore, the oxygen plasma treatment on the surface of as-deposited $BaTa_2O_6$ resulted in a lowering of the interface barrier height and thus, a reduction of the leakage current at Al under a negative bias. This can be explained by the formation of Ba-rich metallic layer by surface etching effect and by filling the oxygen vacancies in the bulk.**

***Index Terms*—Dielectric thin films, optoelectronic display, sputtered films, conduction mechanism.**

I. INTRODUCTION

PREPARATION of thin films with high dielectric constant, low leakage current, and high dielectric breakdown strength is important for the development of the low-voltage driven thin film electroluminescent device (TFEL) [1]. Many studies in this field have attempted to utilize the high dielectric constant, low leakage and high breakdown properties of the amorphous Ta_2O_5 films and other perovskite thin films [2]–[8]. In the case of TFEL, the stacked capacitor structure has been proposed to obtain a reliable EL device with long life time as well as to reduce the power consumption due to the voltage drop in dielectric layers of the EL structure [9], [10]. Particularly, the dielectric materials formed on indium-tin-oxide (ITO) coated glass were intensively investigated in order to use them as an insulating layer for the battery-driven portable TFEL device. In order to manufacture TFEL device profitably, it is necessary to produce devices with an excellent dielectric behavior because the dielectrics must operate reliably withstanding extraordinarily high electric fields over the large area of the glass panel [11]. The success of the display manufacturing can be crucially dependent in part on the ability to produce good-quality defect-free dielectric films over large areas. A number of dielectric systems have been tested with the idea of reducing the cost and improving the display performance, specifically lowering the threshold voltage for applications such as large area displays and extremely small head-mounted displays (HMD). Most of them utilize Ta_2O_5 as one component of the multilayer dielectric system because of its reasonably high dielectric constant. However, the usage of Ta_2O_5 has a number of limitations in color EL device applications because of the numerous wet or dry etching processes of phosphor layer and also, the easy reaction with ITO. Ta_2O_5 easily changes its phase and crystallizes at high temperatures, too. The refractive index of Ta_2O_5 matches well to that of ZnS host, but interface states are generally shallow. These shallow interface states have been noticed by the soft turn-on of the transferred charge versus applied voltage and the luminance (L) versus applied voltage (V) characteristics.

There have been few reports on the operating characteristics of TFEL devices with $BaTa_2O_6$ (BTO) as a dielectric layer [13]–[15]. This paper investigates the electrical characteristics of $BaTa_2O_6$ thin films formed on ITO-coated glass by rf-magnetron sputtering at temperature lower than 200 °C and furthermore, examined the effect of the post-deposition process on the surface and the bulk properties. The conduction characteristics of the ITO/BTO/Al structure will be reported by us for the first time in this work.

II. EXPERIMENTS

All BTO films were prepared by the rf-magnetron sputtering technique on the corning 7059 glass substrates coated with ITO with a sheet resistance of about 20 Ω/□. A 4-in $BaTa_2O_6$ ceramic target with 99.99% purity was used. The base pressure in the chamber was below 5×10^{-5} torr and the pressure during the deposition was maintained at 5 mtorr of Ar and O_2 (20%) gas mixture. The optimum substrate temperature T was determined and maintained at 200°C unless specified and the thickness was varied from 200 to 400 nm in order to examine the bulk effect. To investigate the effect of the post-deposition treatment on the electrical characteristics of the as-deposited films, a part of specimens was exposed to the oxygen plasma for 5 min using the reactive ion etcher.

The top Al electrodes of 0.7 mm in diameter were formed onto the BTO film by thermal evaporation and then the electrical

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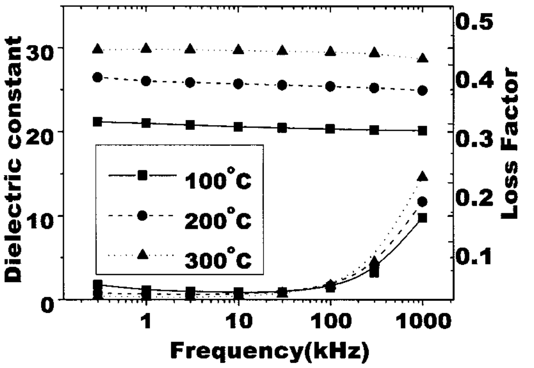


Fig. 1. Frequency dependence of dielectric properties of amorphous $BaTa_2O_6$ thin films.

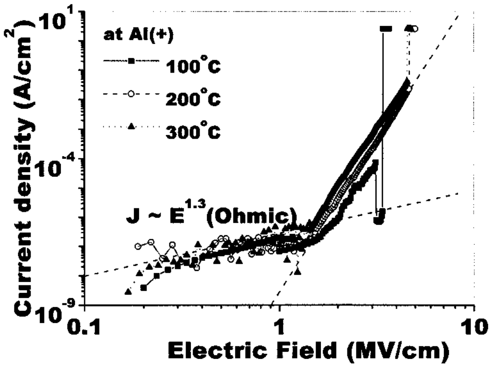
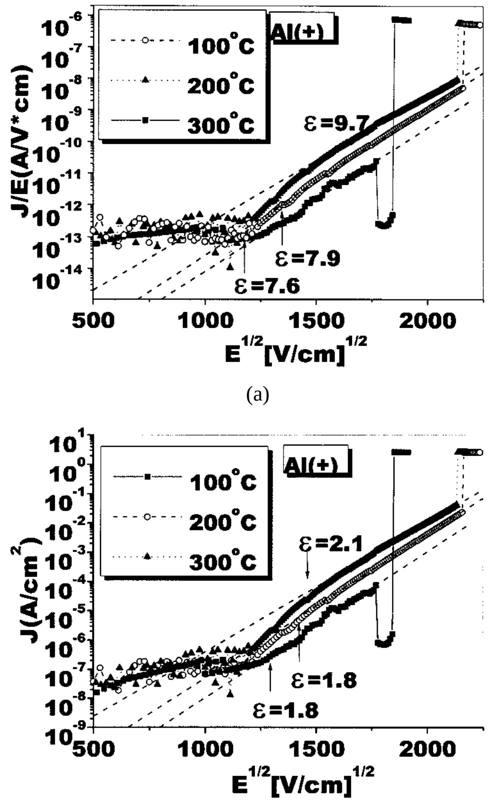


Fig. 2. Leakage characteristics of a series of amorphous $BaTa_2O_6$ thin films with substrate temperature from 100 to 300 °C.



(a)

(b)

Fig. 3. High field conduction characteristic of $BaTa_2O_6$ thin films: (a) $\log(J/E)$ versus $E^{1/2}$ plot and (b) $\ln(J)$ versus $E^{1/2}$ plot.

properties of the capacitors with metal-insulator-metal structure (Glass/ITO/BTO/Al) were characterized. The crystallinity of the thin film was studied by X-ray diffraction measurement (Rikagu, D/Max-3A). The current–voltage characteristics were measured with a fully automated Keithley 237 high-voltage source with the measuring unit at temperatures ranging from room temperature (RT) to 180 °C using a convection oven.

III. RESULTS

A. Electrical Characteristics of BTO Thin Films

Fig. 1 presents typical dielectric properties as a function of applied frequency for the amorphous BTO films prepared by the sputtering technique. The dielectric constant is in the range of 20–30, depending on the substrate temperature during deposition. The dc current versus voltage characteristics were measured with the voltage step of 0.5 V/sec under the negative polarity, i.e., the negative potential was applied to the bottom ITO electrode. In Fig. 2, one can distinguish a step-like structure followed by a high-field region of a relatively fast current increase. This structure of the I–V curve was observed in all BTO films involved in this work. At very low fields, the current density (J) increases linearly with the voltage, i.e., the films display nearly ohmic behavior. The nonlinear behavior at the electric fields exceeding 1 MV/cm can be governed by the space-charge-limited-conduction, the Schottky emission, the Poole–Frenkel (P–F) emission, or tunneling.

At the field of the order of 1 MV/cm or more, the P–F emission and the Schottky emission will be assumed to the most obvious mechanisms inferring from the linearity of slopes in plotting of Fig. 3(a) and (b). In the former case, which is essentially a thermionic emission from a metal electrode into the conduction band of dielectric, the charge carriers, with the image force correction taken into account, are thermally excited over an energy barrier at the ITO-BTO interface. This barrier is created by the equilibration of the energies of the charge carriers in ITO and BTO, and its magnitude at zero field is modified by the field-dependent lowering of the barrier height due to the image forces. Since both the Schottky emission and P–F emission are thermally activated processes, it is necessary to investigate both the field and the temperature dependence of the leakage current in order to determine the operating conduction mechanism. The current density according to the Schottky emission is given by [18]

$$J = AT^2 \exp\left(-\frac{q\phi_o}{k_B T}\right) \exp\left(\frac{\beta}{k_B T} E^{1/2}\right) \tag{1}$$

where

ϕ_0 interfacial barrier height;
T absolute temperature;
q electronic charge;
k_B Boltzmann constant;
A effective Richardson's constant.

The β is defined by

$$\beta = \left(\frac{q3}{4\pi\varepsilon_o\varepsilon_r}\right)^{1/2} \tag{2}$$

where ε_o is the permittivity of the free space and ε_r denotes the high frequency dielectric constant, which is a square of the refractive index. In the Schottky emission, the plot of the log (J) as a function of $E^{1/2}$ and $\log(J/T^2)$ versus $1000/T$ should be a straight line. From the slope of this straight line in the log $(J) - E^{1/2}$ plot, i.e., $\beta/k_B T$, the dielectric constant can be determined [19].

On the other hand, the current density according to the P–F emission predicts a field-dependent behavior given as

$$J = C * \exp\left(-\frac{q\phi_o}{k_B T}\right)\exp\left(\frac{\beta_{pF}}{k_B T}E^{1/2}\right) \tag{3}$$

where C is a constant and β_{PF} is defined by

$$\beta_{\mathrm{PF}} = \left(\frac{q^3}{\pi\varepsilon_0\varepsilon_r}\right)^{1/2}. \tag{4}$$

The conduction mechanism can be determined by comparing the dielectric constant determined from the slope (β or β_{PF}) of the J–V curve with the directly measured value by other methods such as the optical transmission spectroscopy or the capacitance spectroscopy. As described above, the dielectric constant of the insulator in the optical frequency region satisfies the relation of $n^2 = \varepsilon_r$, where n is the optical refractive index [20], [21]. We measured the optical transmission spectra for the film, then calculated the optical refractive index using the Manifacier method [22]. The obtained optical refractive index is 1.6 and the dielectric constant is 2.6 in the optical frequency region. The large deviation of the dielectric constant from the value based on the P–F emission cannot explain the observed J–V characteristics for BTO films. Therefore, the results above suggest that the Schottky emission is the dominant conduction mechanism in the amorphous BTO films in the high field region.

In order to make sure the interface limited conduction, the current dependence of the electric field upon the film thickness should be examined [23]. Fig. 4 indicates the fact that the J–V behavior is independent of the thickness, as expected. Furthermore, since there is an asymmetry in the J–V curves for both polarities of the applied voltage, it is reconfirmed that the conduction is limited by the interface (or electrode). In our case, the asymmetry was attributed to the difference in the work functions of the top and bottom electrode materials.

The results above suggest that there are two dominant conduction mechanisms for the $BaTa_2O_6$ films in the ITO-BTO-Al structure depending on the magnitude of the electric field; namely an ohmic type at low field region, but at higher fields the conduction is governed not by the field enhanced thermal excitation of charge carries from bulk traps but by the Schottky emission as a result of the barrier lowering due to the applied field and the image force.

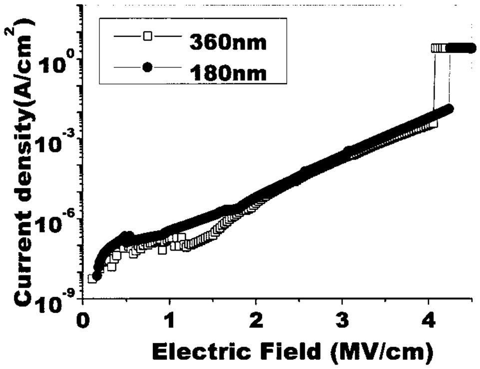


Fig. 4. Thickness dependence of the I–V characteristics for $BaTa_2O_6$ thin films.

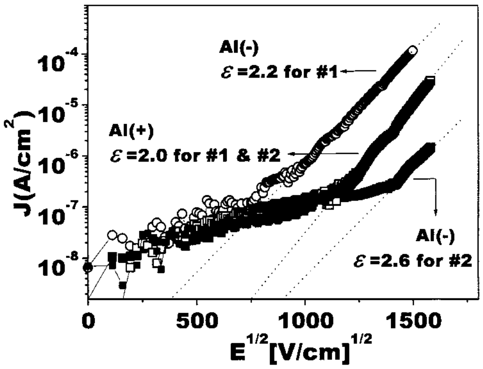


Fig. 5. Polarity dependence of the J versus $E^{1/2}$ curves for as-deposited (#1) and oxygen plasma treated (#2) $BaTa_2O_6$ thin films.

B. Change of Interfacial Characteristics by the Oxygen Plasma-Treatment for As-Deposited BTO Films

The current dominated by the Schottky emission depends on the barrier height between metal-BTO and the barrier height (ϕ_b) depends on the work function of the metal, the barrier lowering by the image force, the surface trap densities, and the surface morphology [24]. Although the as-deposited BTO films have low leakage current characteristics in the amorphous phase, it is necessary to study the post-treatment effect considering the fact that the oxygen vacancies in similar oxide films such as Ta_2O_5 films play a significant role in altering the leakage current behavior [25]. In this work, we investigated the conduction mechanism and furthermore, the possibility of the modification of the interface-limited conduction properties via oxygen plasma exposure.

A part of the as-deposited BTO films was treated under oxygen plasma in a reactive ion etcher and the exposure was performed with a pressure of 72 mtorr for 5 min.

Fig. 5 shows the J–$E^{1/2}$ plots of the as-deposited (#1) and the oxygen-plasma treated (#2) BTO films. The linearity of curves indicates that the conduction is dominated by the Schottky emission or the P–F emission. Above 1 MV/cm, the polarity dependence of the current behavior in #2 is more severe than that of the as-deposited films as shown in the Fig. 5 and the decrease of the leakage current level is also seen. In the case of Al(+), i.e., electrons are emitted from the ITO side, the current characteristics of both #1 and #2 films are nearly same, indicating that the

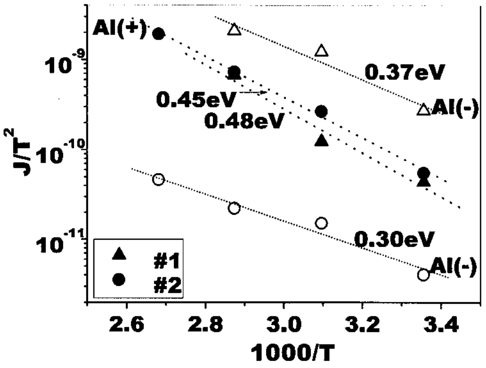


Fig. 6. Plot of $\log(J/T^2)$ versus $1000/T$ at 3 MV/cm for as-deposited and oxygen plasma treated $BaTa_2O_6$ thin films.

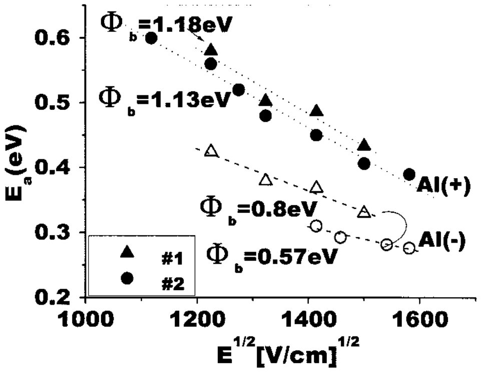


Fig. 7. Polarity dependence of the E_a versus $E^{1/2}$ curve for as-deposited (#1) and oxygen plasma treatment (#2) $BaTa_2O_6$ thin films.

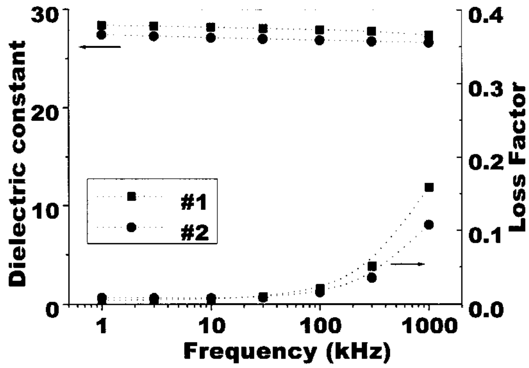


Fig. 8. Effect of oxygen plasma treatment on the C–F characteristics of $BaTa_2O_6$ thin films with 400 nm thickness.

bottom interface is not influenced by the oxygen plasma treatment. The J–V curves for both films show a linear behavior in the log (J) versus $E^{1/2}$ plot above 1.5 MV/cm. When the Al side was negatively biased, the current for the as-deposited films rapidly increased above 0.7 MV/cm whereas an increase of the leakage current for the #2 films was observed above 2 MV/cm. The slope of the curve is given by the (2) and from those plots the values of ε_r were found to be as 2.2 for #1 and 2.6 for #2 films. The magnitude of the dielectric constant was slightly increased by the oxygen plasma exposure possibly due to the increase of O fraction per Ba. Considering the fact that the filling of oxygen vacancies in the perovskite $BaTiO_3$ increases the unit cell size, hence enhancing the polarization, we suggest that the increase of the dielectric constant of #2 can be understood based on the same explanation. One noticeable feature in the curve is the same leakage current level, implying the same conduction mechanism at low fields for both films.

In order to investigate the effect of the post-treatment for #1, the temperature dependent J–V were studied and the Fig. 6(a) shows a variation of the $\log(J/T^2)$ at an applied field of 3 MV/cm as a function of inverse temperature in the form of the Schottky emission (1). The activation energy at the electric field are calculated from the slopes in Fig. 6 and they are 0.48 and 0.45 eV for the #1 and #2 films, respectively. In contrast to this, if the Al side was negatively biased, the activation energy decreased to 0.37 eV for #1 and 0.30 eV for #2 film. The interfacial potential barrier (Φ_b) were determined by extrapolating the plots to $V = 0$ and they are illustrated at Fig. 7. The Φ_b at Al and BTO interfaces are about 0.80 and 0.57 eV for the #1 and #2, respectively. Therefore, we believe that this decrease in the interfacial barrier height with oxygen plasma treatment is due to a change in the band alignment, perhaps due to a slight change in the interface charge state or the strain at the Al-BTO interface.

C. Influence of the Surface Treatment on the Bulk Properties of the BTO

The above results indicate that the surface characteristics between top Al and BTO films can be changed by the oxygen plasma treatment and, furthermore, the barrier height for the electron injection can be lowered. The barrier height between the plasma-treated BTO and the Al electrode is lower than that between the as-deposited BTO and Al. This implies that the low barrier height tends to increase the leakage current from the Al electrode to the BTO films, and as a result the E_{bd} of the BTO will be lowered. Here, note that the lowered barrier height after an exposure to the plasma is not consistent with the decrease in leakage current level in high field region as shown in Fig. 5. This result is possible only if the conduction is affected by the bulk-limited conduction, although the quantitative evaluation is not possible at this moment.

In order to understand the reduction of the leakage current level, but at the same time to understand the lowering of the interfacial barrier height observed in #2 films, we considered the decrease of the surface roughness after the plasma treatment as one of the possible causes. Contrary to the expectation, the root-mean-square roughness probed by the atomic force microscopy from the 10 μm × 10 μm scan was 1.05 and 1.04 nm for #1 and #2 films, respectively, indicating the lack of significant changes in the surface roughness. So, the surface morphology is not a determinant parameter for the leakage current level for #2 films.

On the other hand, the chemical profiles of BTO layer near the surface examined by auger electron spectroscopy depth profiling showed a little variation with the treatments. Since the spectra showed nearly the same carbon contamination for the surfaces of both films, it is imaginable that the reduction of the leakage current densities may come from the composition changes in our films. Especially, the lowered interfacial barrier height at Al-$BaTa_2O_6$ (#2) films could be resulted from the Ba-rich surface induced by the slight etching effect during the plasma exposure. The as-deposited $BaTa_2O_6$ was found to be

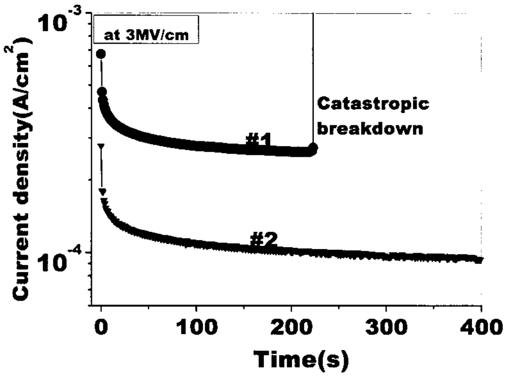
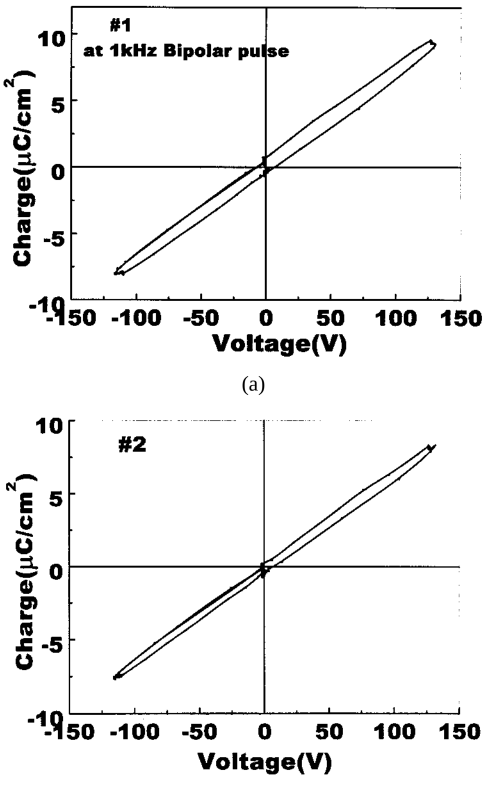


Fig. 9. Effect of oxygen plasma treatment on the I–T characteristics of BaTa$_2$O$_6$ thin films with 400 nm thickness.



(a)

(b)

Fig. 10. Effect of oxygen plasma treatment on the Q–V characteristics of (a) as-deposited and (b) oxygen plasma treated BaTa$_2$O$_6$ thin films.

Ta-rich and Ba deficient near the surface, as compared to the bulk. The oxygen plasma treatment resulted in an increase of the surface Ba : Ta ratio by nearly two times and an increase of the bulk oxygen concentration. The increase of oxygen content may be one of the causes for the decrease of the dielectric loss in Fig. 8.

Results for the as-deposited and the plasma-treated films recorded with time under the field of 3 MV/cm showed the time-dependent variation of the leakage current as shown in Fig. 9. Under the steady-state condition, the as-deposited films show a higher leakage current level as well as a catastrophic breakdown after about 200 s whereas #2 films withstand two times longer than that of the as-deposited films.

Finally, we measured the transferred charge (Q) versus bipolar pulse voltage (V_{peak}) characteristics, as shown in Fig. 10, to examine the applicability of our films to the TFEL devices under the bipolar pulse field of 3 MV/cm. The loop represents the leakage charge during the stressing and the maximum leakage charge is 1.2 and 0.62 μC/cm^2 for the #1 and #2, respectively, indicating a significant reduction of leakage charge in the case of applying the post plasma process.

Therefore, it is clear that the oxygen plasma treatment is very effective for improving the interface properties like the barrier height as well as the bulk characteristics of the leakage current, implying that our BTO films are suitable for the dielectrics of TFEL devices.

IV. CONCLUSION

A study on the temperature dependent current versus voltage characteristics demonstrated a clear evidence of two stage conduction mechanisms. For ITO-BTO-Al structure formed on glass, the high field conduction is dominated by the Schottky emission. Furthermore, the interfacial barrier height was lowered to 0.57 eV in the plasma-treated films compared to 0.80 eV of the as-deposited film, accompanying a reduction of the leakage current level. We believe that the decrease in the barrier height after the oxygen plasma treatment is due to a formation of Ba-rich metallic layer in the film by slight etching effect. Also, the reduction of leakage current level is understood considering that the filling oxygen vacancies, which exist in some depth from the top surface of BTO, effectively by energetic oxygen plasma. This study gives the first insight into the electronic conduction mechanism in the BTO films and shows that the interfacial characteristics can be modified by the post-plasma treatments, accompanying improvement of bulk properties. Analysis of the TFEL devices with the BTO films is in progress and will provide a better understanding of the device physics for the TFEL.

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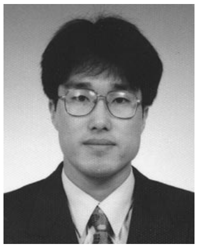
Dr. Lee is a member of the Society for Information Display (SID), the Korean Physical Society and the Korea Institute of Electrical Engineering (KIEE).



Young-Sik Kim was born in Pusan, Korea. He received the B.S. and M.S. degrees from Korea University, Seoul, in 1991 and 1993, respectively. He is currently pursuing the Ph.D. degree in department of electrical engineering at Korea University.

From 1993 to 1996, he worked at Hyundai Electronics, Kyungki-do, as a SDRAM Circuit Engineer. In 1998, he was a Student Researcher at Korea Institute of Science and Technology (KIST), Seoul.

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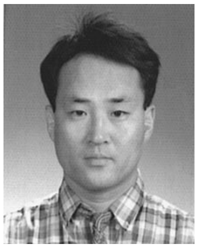
Dr. Kim is a member of the American Physical Society, the Korean Physical Society and the Korean Sensors Society.



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