Structural and Electrical Properties of Bismuth Magnesium Niobate Thin Films deposited at Various Temperatures

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Structural and electrical properties of the fully crystallized-bismuth magnesium niobate ($Bi_2Mg_{2/3}Nb_{4/3}O_7$, BMN) films with 15 mol% excess bismuth deposited on Pt bottom electrode by pulsed laser deposition are characterized for various deposition temperatures. The BMN films were crystallized with a monoclinic structure from 300 °C and the surface roughness slightly decreases with increasing deposition temperature. The capacitance density of the films increases with increasing deposition temperature and especially, films deposited at 400 °C exhibit a capacitance density of approximately 620 nF/cm^2. The crystallized BMN films with approximately 170 nm thickness exhibit breakdown strength above 600 kV/cm (≤ 10 V) irrespective of deposition temperature and a leakage current density of approximately 2 x 10^{-8} A/cm^2 at 590 kV/cm (at 10 V).

Keywords : BMN thin films, Deposition temperature, PLD process, Leakage current density, Dielectric properties

1. INTRODUCTION

Among the embedded passive components, capacitors are most widely studied because they are the major components in terms of size and number. The efforts for embedding materials with medium-to-low permittivity into laminate packages are ongoing. In order to perform all-high temperature processing steps required by the oxide dielectric before the embedding process, film deposition was achieved on metal foils that could be subsequently embedded. Zou et al.[1] demonstrated lead zirconate titanate (PZT) deposited on stainless-steel and titanium foils. Maria et al.[2] also prepared PLZT capacitors on electroless nickel on copper foil prior to embedding and demonstrated capacitance densities (C/A) of 300-400 nF/cm^2. Ferroelectric films were annealed at 500 ~ 600 °C to obtain a high capacitance density after deposition at room temperature by metalorganic decomposition (MOD) process.

BMN has a pyrochlore structure in nature and is known to have high dielectric constants of ~ 210 with a low dielectric loss tangent of 3 x 10^{-4} in bulk materials[3,4]. BMN films for embedded capacitor applications on copper clad laminate (CCL) substrates were studied below a deposition temperature of 150 °C by Park et al.[5-7]. In this case, the films deposited at a low temperature include the partially crystallized phases. An investigation of their structural and electrical properties in fully crystallized BMN films at high temperature was required for embedded capacitor applications. In this study, the crystallized films were prepared on $Pt/TiO_2/SiO_2/Si$ substrates at various deposition temperatures by pulsed laser deposition. Effect of deposition temperature on dielectric and electrical properties was investigated in fully crystallized BMN films with 15 mol% excess bismuth.

2. EXPERIMENTAL

BMN thin films were grown at various temperatures by pulsed laser deposition (PLD) using a pulsed KrF excimer laser (248 nm, Lamda Physik COMPexPro 201). A BMN of 1 inch diameter was used as a target. The base pressure of the chamber used was approximately 5.3 x 10^{-4} Pa. The laser density and repetition rate was 1.5 J/cm^2 and 4 Hz, respectively. Oxygen was added into the chamber during deposition and the oxygen pressure was maintained at 30 mTorr. Thin films were deposited on $Pt/TiO_2/SiO_2/Si$ substrates at various deposition temperatures. The bismuth concentration in the films was obtained using ceramic target having 15 mol% excess bismuth content. The bismuth concentration was confirmed by Rutherford backscattering spectroscopy in the films deposited at various temperatures. Film thicknesses were maintained at approximately 170 nm in the films deposited at various temperatures and were identified through the cross-sectional images by scanning electron microscopy (SEM). Surface morphologies and the crystalline phases of the films were characterized by atomic force microscopy (AFM) and X-ray diffraction (XRD), respectively. For electrical measurement, Pt top electrodes with dimensions of 100 x 100 μm^2 were exactly patterned by lift-off lithography and sputtered by dc magnetron sputtering. After deposition of Pt top electrode, treatment of top electrode was performed at 400 °C for 5 min in oxygen ambient. The dielectric properties of Pt/BMN/Pt/TiO_2/SiO_2/Si capacitors were evaluated by impedance analysis (HP 4194A). The leakage current characteristics of MIM (metal-insulator-metal) capacitors were investigated by HP4145B semiconductor parameter analysis.

3. RESULTS AND DISCUSSION

Figure 1(a) shows X-ray diffraction patterns of BMN films with 15 mol% excess bismuth as a function of deposition temperature. Films deposited down to 200 °C exhibit amorphous-like structure and the fully crystallized phases are observed from 300 °C. From the narrow scanned XRD patterns shown in Fig. 1(b), films deposited at 300 °C show a monoclinic phase of (221) plane alone. On the other hand, films deposited at 400 °C exhibit a typical monoclinic phase with (221) and (004) planes, indicating the strong peak intensity of (004) plane. An increase of peak intensity with increasing deposition temperature produces an increase of grain size from the full-width half maximum (FWHM) values of (221) plane. Accordingly, an increase of grain size impacts on an increase of the capacitance density (dielectric constant) of the films.

Figure 2 shows the variations in rms (root mean square) roughness of the films as a function of deposition temperature. The rms roughness slightly decreases with increasing deposition temperature, indicating the dense morphologies with increasing deposition temperature. In order to observe the surface roughness distinctly, the three-dimensional AFM images of the films deposited at 100 and 400 °C also were shown in an inset of Fig. 2. From the surface images, surface roughness of the films deposited at 100 and 400 °C did not show a big difference from 7.5 to 6.5 Å.

Figure 3 shows the variations in capacitance density and dissipation factor of the films as a function of deposition temperature. The capacitance density of the films slightly increases with increasing deposition temperature and the films deposited at 400 °C show an abrupt increase of capacitance density. An increase of grain size with increasing deposition temperature produces

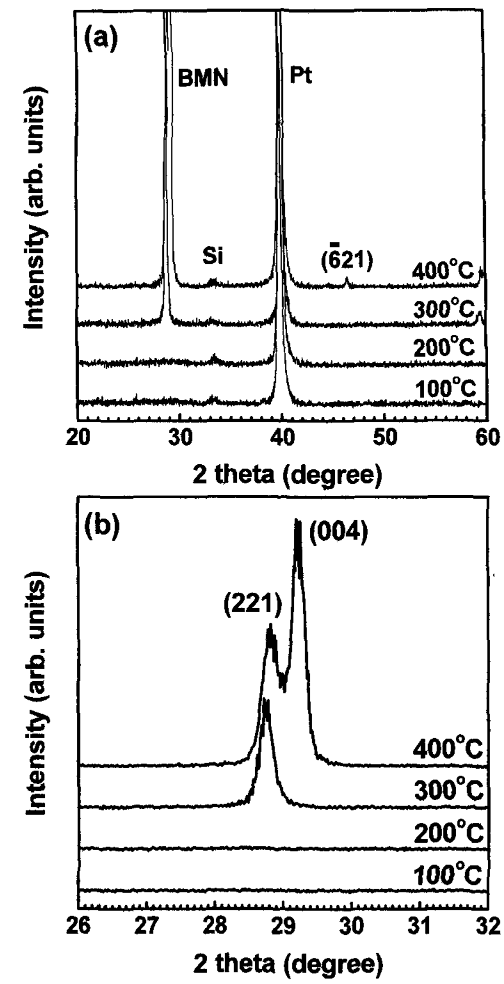

Fig. 1. (a) X-ray diffraction patterns of BMN films with 15 mol% excess bismuth as a function of deposition temperature and (b) the narrow scan of BMN peaks exhibiting at near $2\theta = 29°$ at various deposition temperatures.

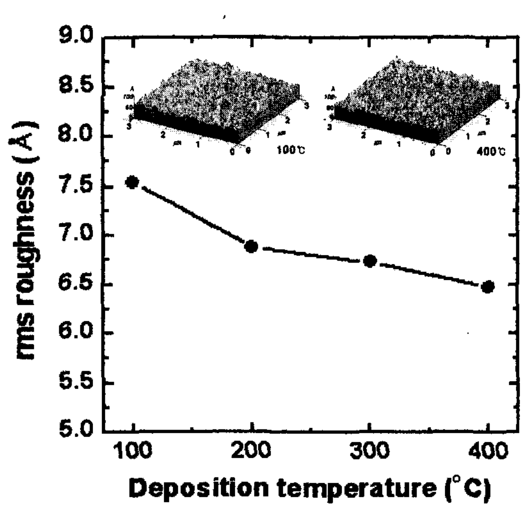

Fig. 2. Variations in rms roughness of the films as a function of deposition temperature. The inset shows three-dimensional AFM images of the films deposited at 100 °C and 400 °C.

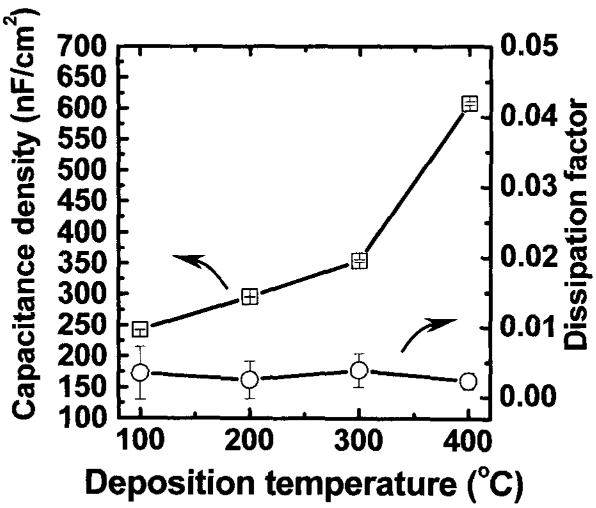

Fig. 3. Capacitance density and dissipation factor of the films as a function of deposition temperature.

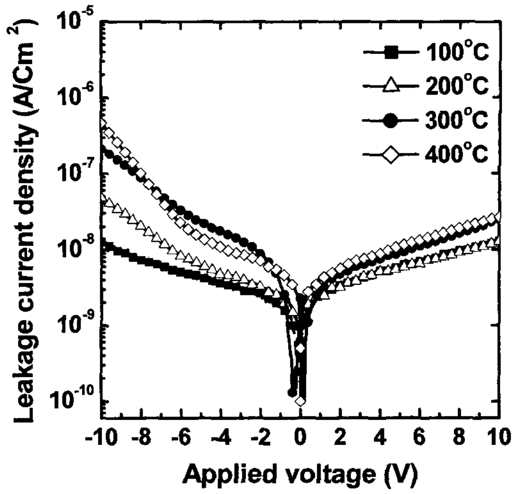

Fig. 4. Leakage current density vs. applied voltage of the films deposited at various temperatures.

an increase of capacitance density (dielectric constant) because an orientational polarization of the dielectric materials increases with increasing the grain size. The capacitance density of the films deposited at 400 °C was approximately 620 nF/cm^2, which is higher than values reported by Maria group[2]. On the other hand, dissipation factor of the films was constantly maintained at 0.3 % irrespective of an increase of deposition temperature.

Figure 4 shows the variations in leakage current density vs. applied voltage of films deposited at various temperatures. For real leakage current density of the crystallized films, Pt top electrode was thermally annealed at 400 °C for 1 min to relieve the charge density formed at interface between Pt and BMN films during deposition of the Pt top electrode. The leakage current densities show asymmetric behavior for negative and positive electric field as shown in Fig. 4. Even though the same Pt electrode was used both top and bottom side, an interface state between bottom Pt and BMN films is different from that between top Pt and BMN films because BMN deposition on bottom Pt takes for a long time at high temperature. The crystallized BMN films with approximately 170 nm thickness exhibit breakdown strength above 600 kV/cm ($\leq$ 10 V) irrespective of deposition temperature and a leakage current density of approximately 2 x 10^{-8} A/cm^2 at 590 kV/cm (at 10 V), as shown in Fig. 4. This result shows a superior leakage current characteristics than that observed by 1 μm-thick PZT thin films[1]. The fully crystallized BMN films are a suitable candidate for embedded capacitor applications compared with the ferroelectric films.

4. CONCLUSION

The BMN films with 15 mol% excess bismuth deposited on Pt bottom electrode by pulsed laser deposition were crystallized with a monoclinic structure from 300 °C and the surface roughness slightly decreases with increasing deposition temperature. The capacitance density of the films increases with increasing deposition temperature and especially, films deposited at 400 °C exhibit a capacitance density of approximately 620 nF/cm^2. The crystallized BMN films with approximately 170 nm thickness exhibit breakdown strength above 600 kV/cm($\leq$ 10 V) irrespective of deposition temperature and a leakage current density of approximately 2 x 10^{-8} A/cm^2 at 590 kV/cm (at 10 V). The fully crystallized BMN films are a suitable candidate for embedded capacitor applications.

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REFERENCES

[1] Q. Zou, H. E. Ruda, B. G. Yacobi, K. Saegusa, and

M. Farrell, "Dielectric properties of lead zirconate titanate thin films deposited on metal foils", Appl. Phys. Lett., Vol. 77, p. 1038, 2000.

[2] J. P. Maria, K. Cheek, S. Streiffer, S. H. Kim, G. Dunn, and A. Kingon, "Lead zirconate titanate thin films on base-metal foils: An approach for embedded high-permittivity passive components", J. Am. Ceram. Soc., Vol. 84, p. 2436, 2001.

[3] D. P. Cann, C. A. Randall, and T. R. Shrout, "Investigation of the dielectric properties of bismuth pyrochlores", Solid State Communications, Vol. 100, p. 529, 1996.

[4] J. C. Nino, H. J. Youn, M. T. Lanagan, and C. A. Randall, "Bi_2O_3 solubility of Bi-based pyrochlores and related phases", J. Mater. Res., Vol. 17, p. 1178, 2002.

[5] J. H. Park, C. J. Xian, N. J. Seong, S. G. Yoon, S. H. Son, H. M. Chung, J. S. Moon, H. J. Jin, S. E. Lee, J. W. Lee, H. D. Kang, and Y. K. Chung, "Bismuth-based pyrochlore thin films deposited at low temperatures for embedded capacitor applications", Jpn. J. of Applied Physics part 1, Vol. 45, p. 7325, 2006.

[6] J. H. Park, C. J. Xian, N. J. Seong, S. G. Yoon, S. H. Son, H. M. Chung, J. S. Moon, H. J. Jin, S. E. Lee, J. W. Lee, H. D. Kang, Y. K. Chung, and Y. S. Oh, "Realization of a high capacitance density in $Bi_2Mg_{2/3}Nb_{4/3}O_7$ pyrochlore thin films deposited directly on polymer substrates for embedded capacitor applications", Appl. Phys. Lett., Vol. 89, p. 232910, 2006.

[7] C. J. Xian, J. H. Park, S. G. Yoon, J. W. Lee, W. C. Kim, S. T. Lim, S. H. Sohn, J. S. Moon, H. M. Jung, S. E. Lee, I. H. Lee, Y. K. Chung, M. K. Jeon, and S. I. Woo, "Effect of thickness on electrical properties of bismuth-magnesium niobate pyrochlore thin films deposited at low temperature", J. Appl. Phys., Vol. 101, p. 084114, 2007.