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Ferroelectric properties of barium titanate thin film prepared from Ti-deposited Si substrate by hydrothermal synthesis

C. R. Cho, M. S. Jang, S. Y. Jeong, and S. J. Lee
Department of Physics, Pusan National University, Pusan, 609-735, R. KOREA

In recent years there has been a surge in research actively on barium titanate thin films for non-volatile memory[1] and ferroelectric dynamic random access memory devices[2] applications. New method and processing techniques[3,4] are being developed for producing device quality films. Generally, Various methods like MOCVD[5-7], sputtering[8,9], and sol-gel[10-12] have been used for the crystallization of $BaTiO_3$ films. These methods demand a post deposition annealing at temperatures above 500℃ for longer than 1h, irrespective of film fabrication method[13-15]. There high temperature processing conditions often cause problems at the film substrate interface[16], and hence, difficulties in integrating the films with silicon monolithic circuits. The hydrothermal method recently is being adopted for the $BaTiO_3$ thin film preparation, but the C-V characteristics and ferroelectric properties of the film has not been measured.[17-19] This synthesis involves the "dissolution-recrystallization" process[20] of a Ti metal in a $Ba(OH)_2$ solution to deposit a crystalline layer of $BaTiO_3$ on the surface. In this work we will present results on a synthesis procedure which includes a buffer layer of SiO_2 beween the Si substrate and the Ti film on which the reaction takes place. The first, we will be report the ferroelectric properties of the polycrystalline $BaTiO_3$ thin film synthesized by hydrothemal reaction.

1.0μm-thick titanium layers were deposited on the boron-doped Si(100) substrate [SiO_2(20nm) was thermally oxidized on the surface] at 300℃ by electrocn beam evaporator. X-ray diffractograms of the films showed that the SiO_2 layers were amorphous and the Ti film polycrystalline (preferentially oriented in the [002] direction). Circular gold dot electrodes of area $2.5x10^{-3}cm^2$ were evaporated on to the $BaTiO_3$ films with the film temperature kept 250℃ in a vacuum of 10^{-5} Torr.

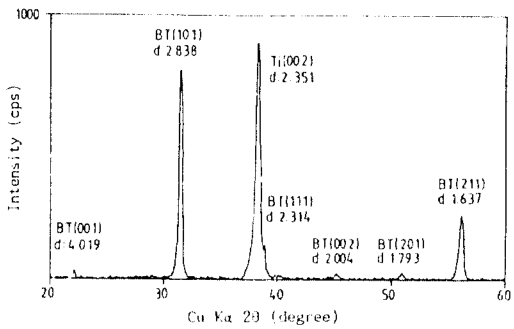

Figure 1

XRD patterns of the thin films processed at 220℃ in 2.0N $Ba(OH)_2$ for 24 h is shown in Fig. 1. Polycrystalline of $BaTiO_3$ thin film showed no preferred orientation. Peak profile was sharp enough to prove the crystallinity of barium titanate, but not enough to identify its tetragonality. The scanning electron micrographs(SEM) of the surface and the fracture surface of are shown in Figure 2(a) and 2(b) show the surface and cross section of the $BaTiO_3$ film formed on SiO_2/Si(100) substrates at 220℃ in 1.0N $Ba(OH)_2$ for 24h by hydrothermal reaction, respectively. The average grain size of the film was estimated to be around 0.5μm and was uniform. Thickness of the thin film are about 600nm. The thickness value is only approximate because the interface between the Ti substrate and $BaTiO_3$ film is not very well defined.

Capacitance-frequency characteristics of 600nm thick synthesized films is shown in Fig. 3. A capacitance of 0.75nF and a tanδ of 0.08 were measured at 10kHz. The increase in capacitance and tanδ at frequencies below 500Hz is due to a contact resistence between the probe and electrode, and no notable frequency dispersion in

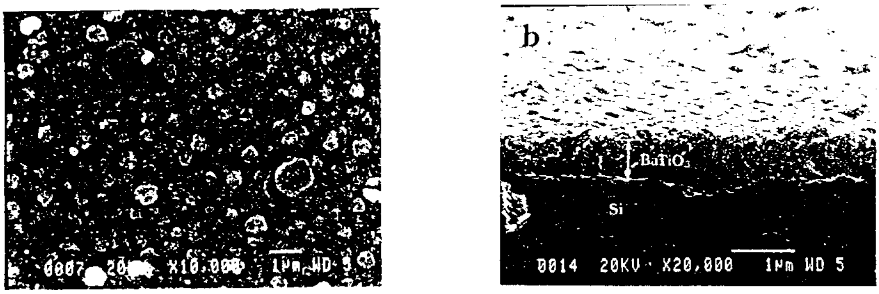

Figure 2

the capacitance was observed in the measurement range. The bias voltage dependence of capacitance(C-V characteristics) was measured for the $BaTiO_3$ thin film capacitors with a MIM structure and shown in Fig. 4. Capacitance was

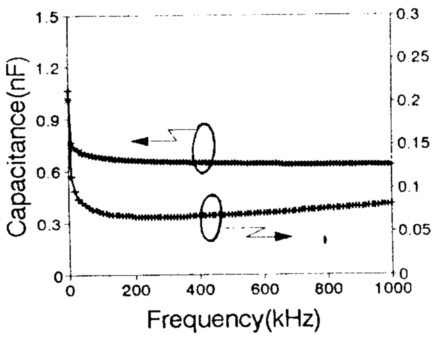

Figure 3

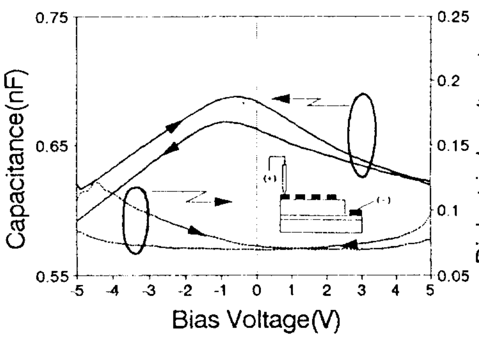

Figure 4

measured at 100kHz with $0.1V_{rms}$ oscillation voltage, using an impedence meter(HP4194A). The magnitude applied bias voltage was adjusted so that the same range of bias field, from -5 to 5V, was applied to dielectric film. The maximum values of the capacitance were taken in about -0.9V bias field. The capacitance decrease and the dielectric loss increase with the bias voltage, which is due to its high leakage, especially at high voltage as shown in figure. The ferroelectric behavior of the barium titanate film was examined by observing the polarization reversal using a Sawyer-Tower circuit. Figure 5 shows a P-E hysteresis loop for a 600nm thick film, measured under an electric field of 20kV/cm at 60Hz. The loops observed in the present investiga-0tions show incomplete saturation. A further increase in the electric field in the film caused its breakdown. The non-saturation loop in the films may be attributed to a complex conduction process due to depolarizing fields and defects at grain boundary and the interface[21]. The saturation naturre of the loop provides strong evidence of the ferroelectric behavioof the film. The measured valuse of remanent polarization(P_r) and coercive field(E_c) are 25μC/cm^2 and 6.5kV/cm respectively. The value of P_r is higher and E_c is lower than those reported for bulk single crystal.

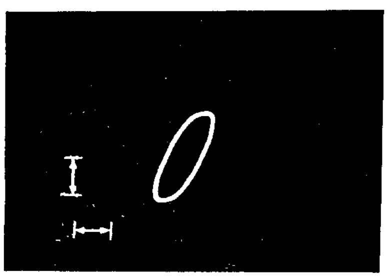

Figure 5