Lead-free Piezoelectric (Na, Bi)TiO_3-$BaTiO_3$ Thin Films and Their Application

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Piezoelectric materials have been widely used for sensors and actuators. Thin films of piezoelectric materials prepared on Si substrates can be processed into any configurations by standard micro-electro-mechanical systems (MEMS) process [1, 2]. Performance enhancement and miniaturization are easier for piezoelectric-MEMS devices than for normal Si-MEMS devices. It is expected that the demand for piezoelectric thin films will increase in the future. We found that Pb(Zr,Ti)O_3 (PZT) thin films with (001) orientation synthesized by rf-magnetron sputtering on Si substrates show excellent piezoelectric properties. Using these piezoelectric thin films, we proposed and developed new types of sensors and actuators of MEMS devices [3]. These sensors and actuators realized higher performance and easier miniaturization than conventional ones. We have successfully turned these piezoelectric-MEMS devices into mass-production of angular rate sensors and ink jet head actuators since 2003 and 2004, respectively.

In recent years, lead-free piezoelectric materials have been intensely studied to address environmental concerns of PZT. Nevertheless definite alternatives have not been found yet. To change this situation, we focused on piezoelectricity in thin films where peculiar properties sometimes appear. A solid solution composed of rhombohedral $(Na_{0.5}Bi_{0.5})TiO_3$ and tetragonal $BaTiO_3$, $(1–x)(Na_{0.5}Bi_{0.5})TiO_3–xBaTiO_3$ (NBT−BT) has been recognized as a promising lead-free piezoelectric material since NBT−BT shows large electric (*E*)-field induced strain at the composition near the morphotropic phase boundary (MPB) of x = 0.06–0.07 [4]. We demonstrated that epitaxial thin films of NBT−BT deposited on MgO single-crystal substrates by rf-magnetron sputtering exhibit very large piezoelectricity comparable to those of the PZT thin films used for sensors and actuators [5-8]. Very recently, we prepared the c-axis oriented polycrystalline thin films of NBT–BT around the MPB on a $LaNiO_3$-buffered Si substrate by rf-magnetron sputtering [9]. Control of the sputtering conditions gave rise to a variety of tetragonality, c/a=1.001~1.020, in the NBT–BT thin films. The NBT–BT thin films with high c/a more than ~1.01 were significantly internally biased, and exhibited a linear piezoelectric response with the piezoelectric coefficient, e^*_{31}, reaching -4.8 C/m^2 in the unipolar excitation and a low dielectric permittivity, ε_r, of about 200. The temperature dependence of dielectric properties revealed that the permittivity maximum temperature, T_m, of the thin film was remarkably enhanced to ~550 °C from the ~300 °C of bulk NBT–BT. As the c/a decreases, the internal bias field was gradually relaxed, and the electromechanical strain was increased together with an increase of ε_r. For the NBT-BT thin film with the lowest c/a of 1.001, the e^*_{31} reached as high as -14.6 C/m^2, which is comparable with those of the PZT thin films on Si.

The presentation will describe preparation and properties of the lead-free piezoelectric NBT-BT thin films. In addition, our recent study on the application to lead-free inkjet head actuator will be reported.

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