Preparation of para electric Barium Titanate ceramics by Mn-Nb co-doping for DC-bias free dielectrics

University of Yamanashi [1], °Piyush Sapkota[1], Ichiro Fujii[1], Shintaro Ueno[1], Satoshi Wada[1,*]

[*]E-mail: swada@yamanashi.ac.jp

Owing to the large dielectric constant (ε_r), barium titanate (BT) has been widely used in low electric power electronic devices such as multi-layer ceramic capacitors. However, the strong dependence of ε_r of BT ceramics on the applied electric field has been repeatedly reported, which restricts the use of BT ceramics in high power applications. In this study, Mn-Nb doped BT ceramics were prepared by solid state synthesis and their electrical properties were investigated.

The raw materials used were $BaCO_3$, TiO_2, MnO_2 and Nb_2O_5. Equimolar amounts of Mn and Nb were doped in BT ceramics with three different routes. In the first route, 5 mol % (BTMN 5 TI) and 10 mol % (BTMN 10 TI) Mn and Nb were doped internally as a substituent in the B site of BT ceramics in two steps. Firstly, individual doping of Mn was carried out during first calcination. It was then followed by equimolar Nb doping during second calcination to get $BaTi_{(1-2x)}Mn_xNb_xO_3$. In the second route, similar procedure was applied but 5mol % (BTMN 5 TE) and 10 mol % (BTMN 10 TE) Mn and Nb were doped externally as additives keeping the Ba/Ti ratio as unity. In the third route, both 5 mol % (BTMN 5 OI) and 10 mol % (BTMN 10 OI) Mn and Nb were doped internally in one step as substituent in the B site of the BT ceramics to get $BaTi_{(1-2x)}Mn_xNb_xO_3$. Calcination for all the three routes was done at 1000°C for 5 h. Green compacts formed after binder addition (PVB, 3 wt %) and uniaxial pressing (250 MPa) were sintered conventionally for 5 h at 1300°C. After cutting and polishing into a plate of size 4 mm x 1.5 mm x 0.4 mm, the samples were annealed for 4 h at 1100°C. Au electrodes were deposited on both sides of the plates by sputtering, followed by annealing at 300 °C for 10 min, and then property measurements were done.

XRD patterns, as shown in Fig 1, confirmed single phase perovskite structure with cubic symmetry for BTMN 5 TI and BTMN 10 TI sintered powder samples. Sintered powders of BTMN 5 OI and BTMN 10 OI samples showed secondary phases of hexagonal BT whereas BTMN 5 TE and BTMN 10 TE showed certain unknown phases. Polarisation-electric field loops, as shown in Fig 2, of BTMN 10 TI, BTMN 10 TE and BTMN 10 OI samples resembled para electric nature.

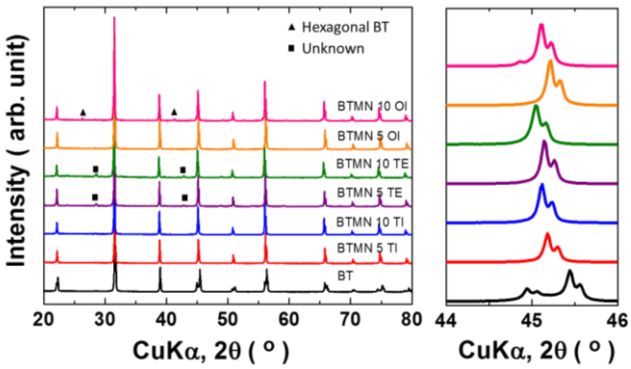

Fig 1. XRD patterns of Mn-Nb co-doped BT ceramics

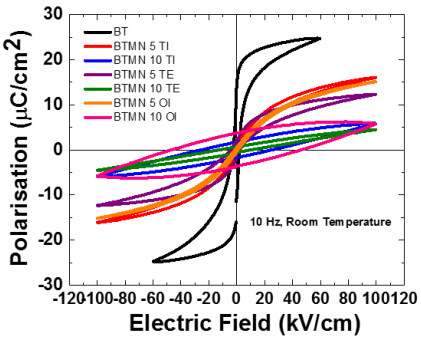

Figure 2, Polarisation-Electric field loops