

Electronic Supplementary Information:

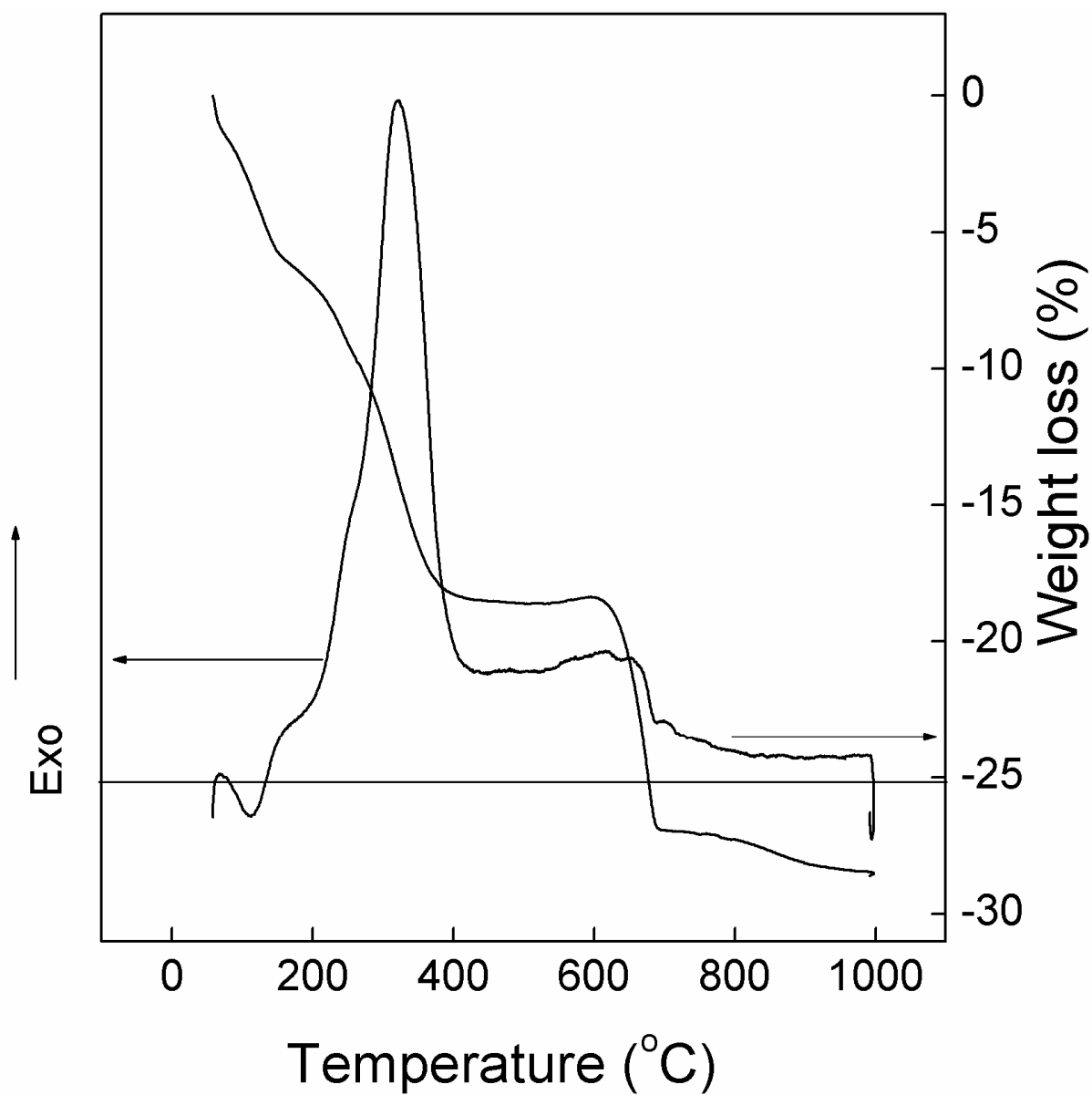


Figure I. Representative DTA- and TGA-plots of a dried microemulsion derived barium titanate precursor powder.

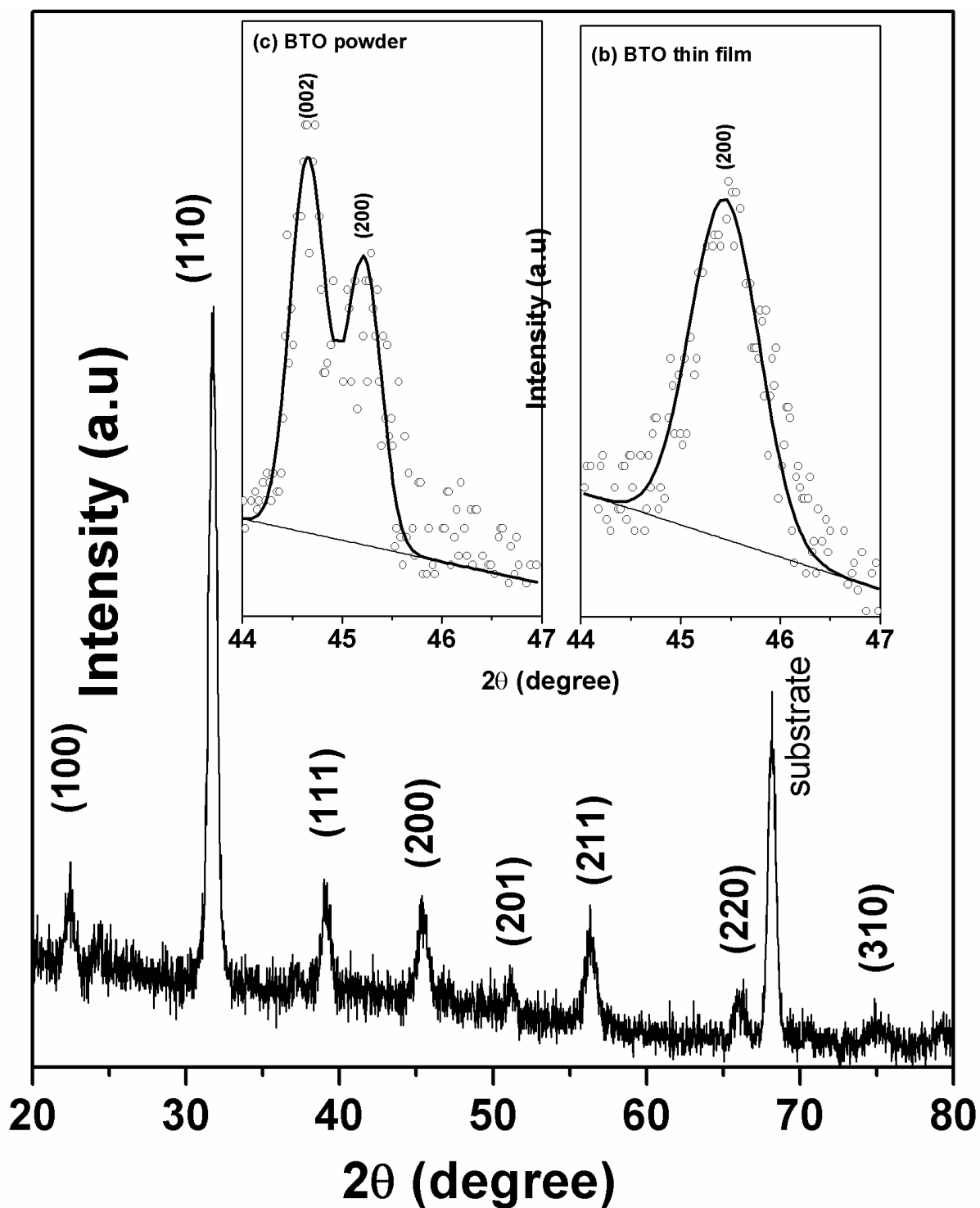


Figure II. X-ray diffraction pattern of barium titanate thin films annealed at 750°C for 10 minutes in oxygen. The insets shows slowly scanned X-ray diffractograms of this film (b) and of a reference powder calcined at 800°C for 2h (c).

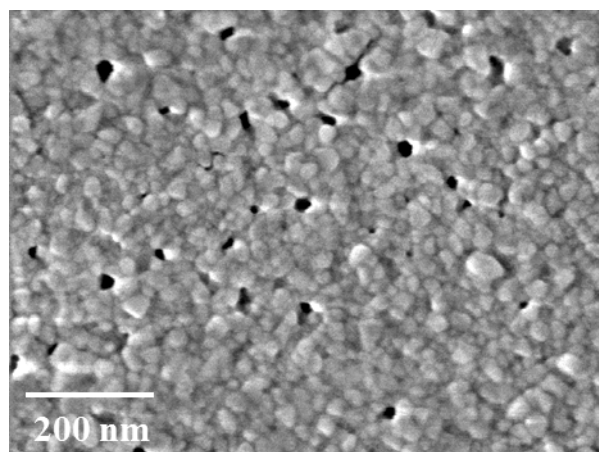


Figure III. FESEM image of the surface morphology of a pure nanoparticle dispersion derived BTO thin film annealed at 750°C for 10 minutes.

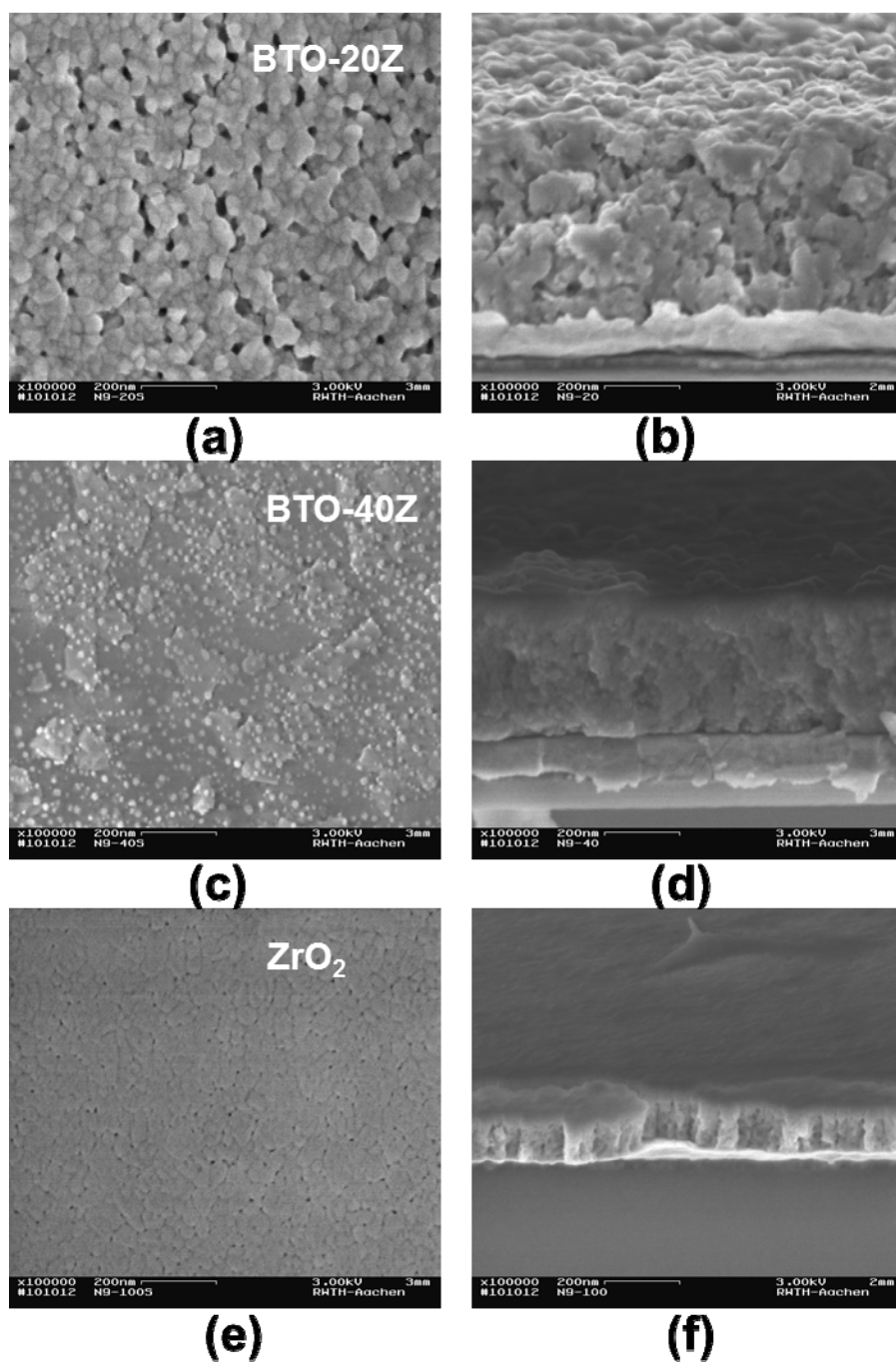


Figure IV. The surface and cross sectional morphology of the BTO-20Z, BTO-40Z, and ZrO₂ films were characterized by FESEM. The surface morphology is shown in the left panel whereas the right panel shows the cross section morphology of BTO-20Z, BTO-40Z, and ZrO₂ thin films, all annealed at 750°C for 10 minutes. Surface porosity is observed in BT up to BT-20Z composite films although the bulk of all these films exhibit a dense microstructure with uniform thickness. The surface porosity could be due to a relatively shorter annealing time (10 minutes) of the topmost deposited layer compared to lower ones, which experience a longer total exposure to the thermal treatment. In these micrographs the nano-crystalline nature of the films can clearly be recognized. The average grain size is approximately 50 nm. We found no significant change of the grain size up to an addition of 20 wt.-% ZrO₂ in the modified BTO films (Figures IV(a) and (b)). However, the surface morphology is quite

different in films with ZrO_2 contents of 30 and 40 wt.-%. In the case of BTO-40Z for example (Figures IV(c and d)) the granular structure of the films disappears and a smoother surface can be observed. The bulk of these films still remains granular in nature. Additionally, these films also exhibit lower porosity. Obviously these films are sintered to yield in a lower porosity even when annealed for a shorter duration, probably due to the higher contents of *con*- ZrO_2 . The film with 40 wt.% ZrO_2 additionally shows some white spherical precipitations on the surface. It seems that ZrO_2 could be phase separated in this film, however, no diffraction peak corresponding to any ZrO_2 polymorph could be identified through X-ray diffraction. Further studies are required to understand this observation. Alkoxy derived pure ZrO_2 films had a dense microstructure with nano-crystalline grain size. As identified in the cross-sectional micrograph a columnar-type grain growth is apparent in ZrO_2 thin films. The interface of all these films (as shown in the cross sectional micrographs) are quite sharp ruling out any inter-diffusion among the constituent elements towards and from the underlying substrates.

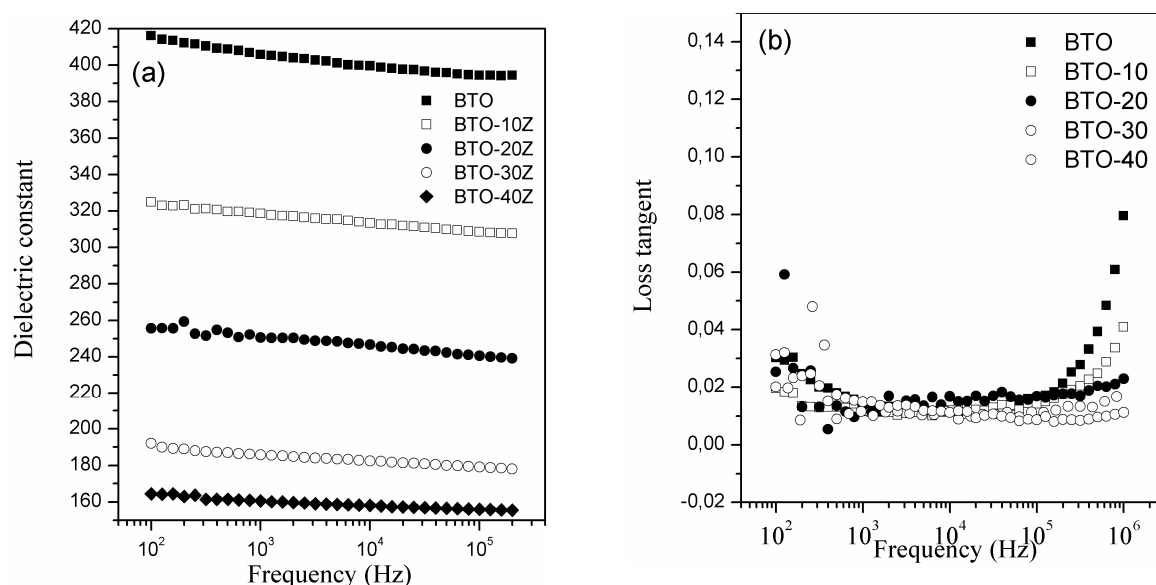


Figure V. Frequency dispersion of (a) the dielectric constant and (b) loss tangents of BTO and BTO- ZrO_2 composite thin films annealed at 750°C for 10 minutes.

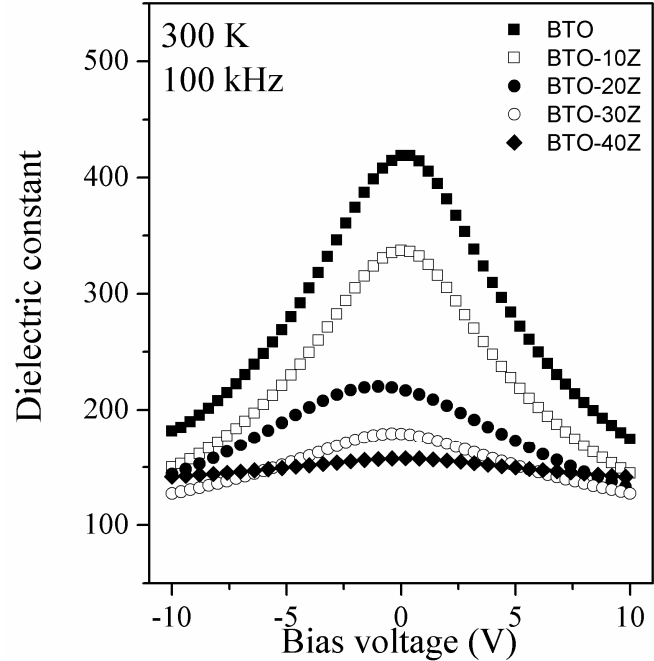


Figure VI. Voltage dependent dielectric constants of BTO and BTO-ZrO₂ composite thin films measured at 300 K and 100 kHz.