Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Flexible $BaTiO_3$/PVDFmultilayernanocomposite film with enhanced dielectric strength and highenergy density

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Electronic supplementary information

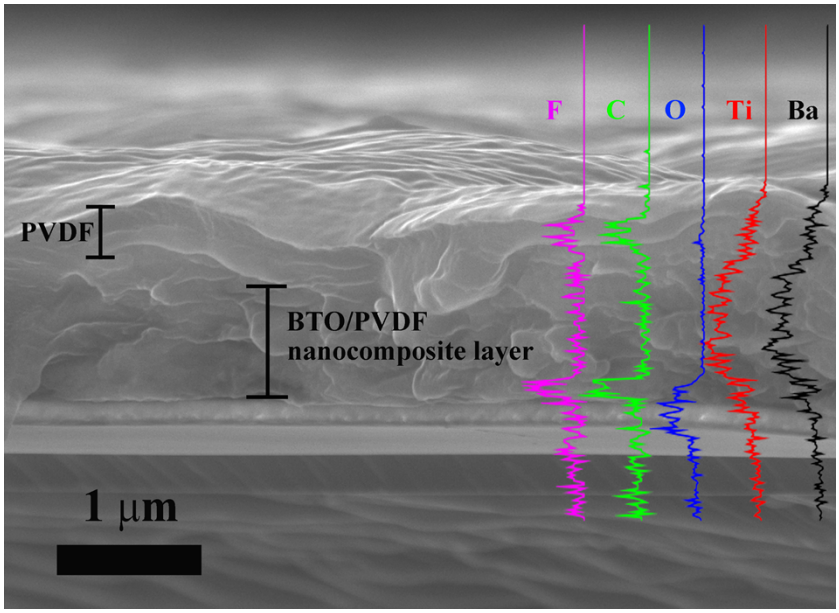

Fig. S1 Cross-sectional SEM image of the BTO/PVDF nanocomposite film (fractured in liquid nitrogen) and EDS line scanning spectrum of this film. From this figure, we can clearly observe the top PVDF layer and the bottom BTO/PVDF nanocomposite layer. The Ba and Ti EDS spectra show that BTO nanoparticles mainly distributed in the bottom nanocomposite layer and have a peak value at the internal layer, confirming the gradation of BTO nanoparticles. Correspondingly, C and F EDS spectra show peak values at the outer layers, which certify the PVDF-rich outer layers.

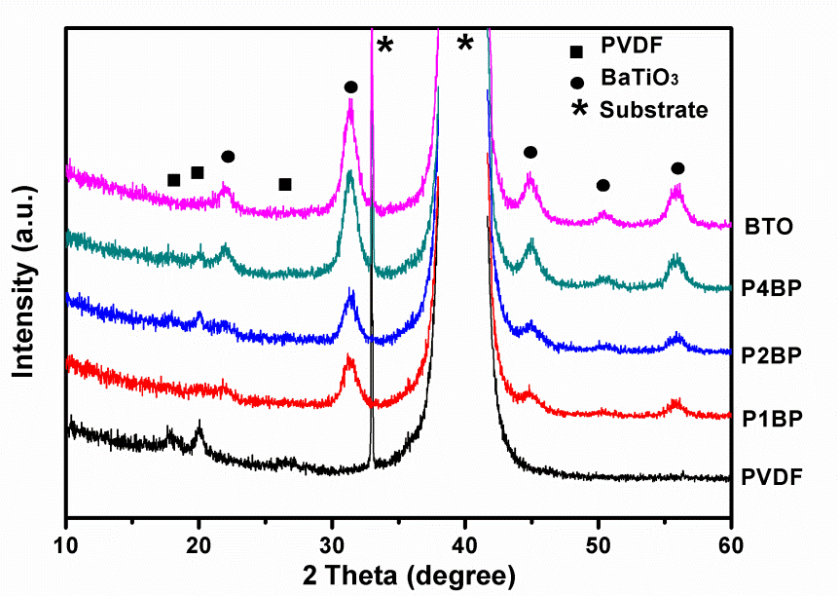

Fig. S2 XRD patterns of the BTO nanocrystal film, sample P1BP, P2BP, P4BP, and the PVDF film.

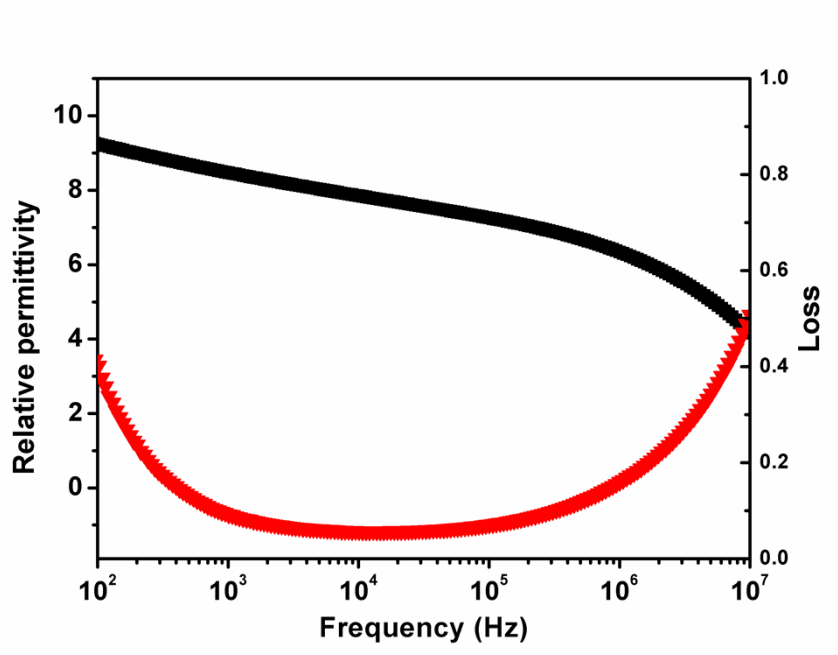

Fig. S3 Frequency-dependent relative permittivity and dielectric loss of the PVDF film. The pure PVDF film has a high dielectric loss at the low frequency and high frequency ends. That's because the PVDF film only has a thickness of 300 nm due to the method. During the sputtering process of the electrode, the thin film was heated and distorted, which caused the permeation of some Pt electrode atoms into the PVDF film and finally resulted in the high dielectric loss at the low frequency end. The dielectric loss at the high frequency end is caused by the glass transition of pure PVDF and the contact quality of the electrode.

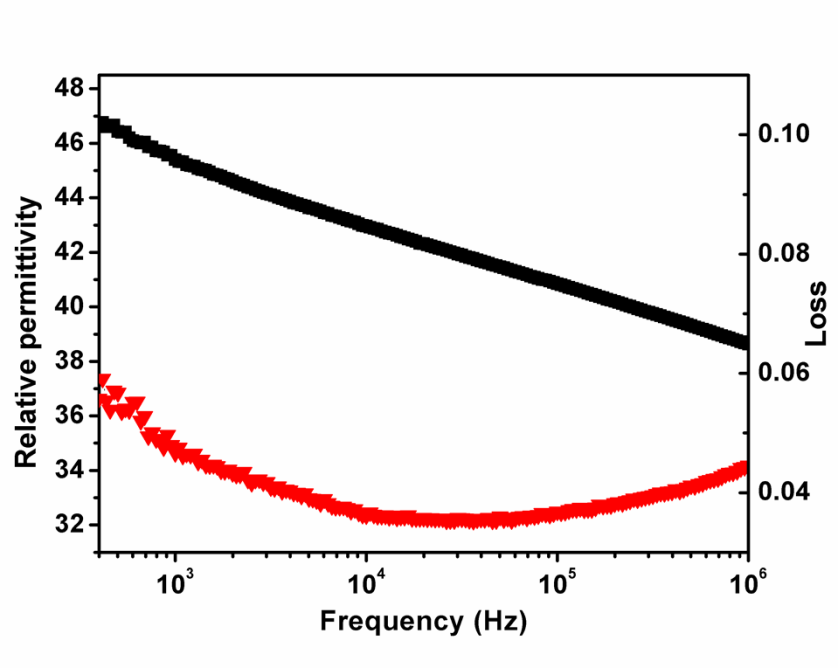

Fig. S4 Frequency-dependent relative permittivity and dielectric loss of a film composed of 8-nm BTO nanocrystals deposited by spin coating dropwise deposition of BTO-TEG suspension (~ 4 layer coating). The nanoparticles were verified by TG analysis to have a 10 wt% TEG organic layer, which make it disperse well in polar solvents. Using the Modified Kerner Model, the BTO-TEG loading is calculated as high as 92.5 vol%. Permittivity values used were 84 for the 8-nm BTO nanoparticles, 37 for TEG and 1 for air.

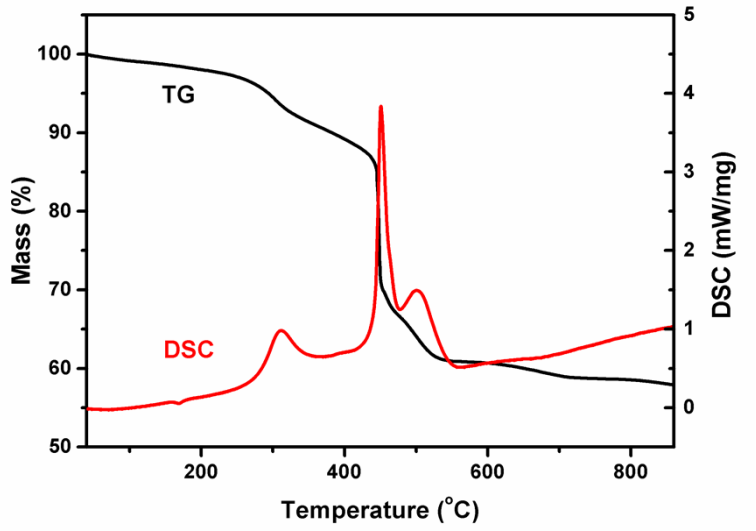

Fig. S5 TG/DSC curve of the "PBP" sample that removed from the substrate. The DSC curve shows three peaks. The peak at ~300 °C corresponds to the decomposition of the surface TEG

organic layer of the BTO nanoparticles. The two peaks at ~500 °C are attributed to the decomposition of the PVDF polymer matrix. From the TG curve, the volume fraction of BTO in "PBP" sample can be calculated to 28.9 vol%, which corresponds well with the volume fraction of ~ 28 vol% that was calculated from the dielectric constant.

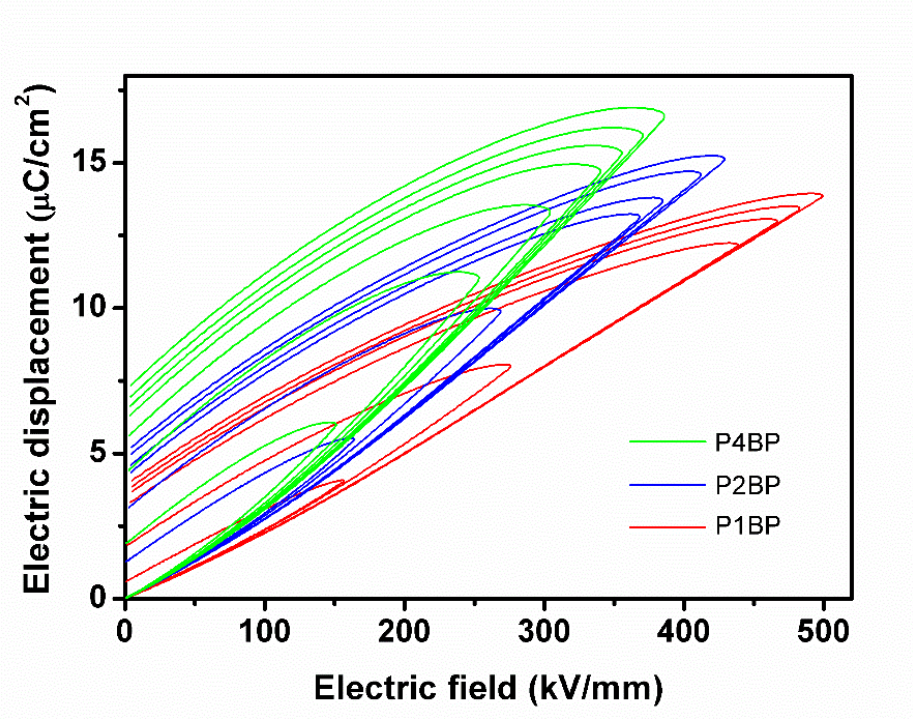

Fig. S6 *D-E* loops of samples P1BP, P2BP and P4BP.