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## **Supplementary Information**

Figure 1S (a-c) is the surface SEM diagram of (1-x)BF-xBT. All samples show a uniform and dense morphology. The average grain sizes gradually decrease from 6.29µm of 0.7BF-0.3BT to 3.09µm of 0.5BF-0.5BT with increasing *x* (Figure 1S (d)). The decreasing grain size is attributed to the high melting point (1618 °C) of BT, which raises the densification sintering temperature, and the disorder in the crystal structure that disrupts the long-range order of the crystal arrangement. Moreover, the Energy Dispersive Spectrometer (EDS) images in Figure 2S confirm the uniform distribution of elements in (1-*x*)BF-*x*BT.



Figure 1S (a-c) Surface SEM, (d) average grain size and relative density for (1-x)BF-xBT (x = 0.3, 0.4, 0.5).



Figure 2S EDS image of (1-x)BF-*x*BT (x = 0.3, 0.4, 0.5).

Figure 3S (a) shows the impedance modulus of (1-x)BF-xBT. The modulus amplitude is only observed at  $0.2 \le x \le 0.4$ , and the amplitude is the largest at 0.7BF-0.3BT. The resonance frequency ( $f_r$ ) and antiresonance frequency ( $f_a$ ) can be determined from the impedance modulus. The electromechanical coupling coefficient ( $k_p$ ) can be calculated by the formula:

$$\frac{1}{k_p^2} = 0.398 \frac{f_r}{f_a - f_r} + 0.579$$

As BT increases,  $k_p$  reaches its maximum value at 0.7BF-0.3BT ( $k_p = 0.28$ ). However, it is quite difficult to calculate the  $k_p$  value when  $x \ge 0.5$  (Figure 3S (b)). This is consistent with variation  $d_{33}$  as a function of x.



Figure 3S (a) impedance modulus and (b)  $k_p$  of (1-x)BF-xBT (x = 0.2 ~ 0.9).