Supplementary information:



Fig. S1. Schematic diagram of the measurement setup.



Fig. S2. The XRD pattern of BCZT powder and the inset is the SEM image of BCZT particles.



Fig. S3. XRD of the BCZT/PDMS composites.



Fig. S4. SEM-BSE images of the cross section of BCZT/PDMS composites: (a) 3 vol.%, (b) 5 vol.%, (c) 8 vol.%, (d) 10 vol.%, (e) 13 vol.% and (f) 18 vol.%.



Fig. S5. SEM images of the BCZT/PDMS composites: (a) aligned and (b) randomly dispersed; SEM images of the BCZT/PDMS composites after adjusting the brightness and contrast: (c) aligned and (d) randomly dispersed; optimal ellipses fitting to the regions of BCZT particles: (e) aligned and (f) randomly dispersed.



Fig. S6. The polarization–electric field (P-E) loops measured at room temperature and 0.2 Hz for the BCZT/PDMS composites.

Composition	8 <sub>r</sub>	$tan\delta$ (%)	<i>d</i> <sub>33</sub> (pC/N)	g <sub>33</sub> (×10 <sup>-3</sup> Vm/N)
3 vol.%	2.8	4.5	2	80.7
5 vol.%	3.6	5.2	3	94.1
8 vol.%	6.6	4.8	6	102.7
10 vol.%	9.0	5.9	7	87.8
13 vol.%	13.0	4.7	9	78.2
18 vol.%	17.0	5.6	12	79.7

Table 1 Electrical properties of BCZT/PDMS composites.