Supplementary Information

All-organic non-percolative dielectric composites with enhanced

electromechanical actuating performances by controlling molecular

interaction

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Figure S1 Comparison maximum actuated strains of ESO/(4 phr PANI/NBR) composites with ESO/(6 phr PANI/NBR) composites filled with different contents of ESO.

The maximum actuated strains of ESO/(4 phr PANI/NBR) composites filled with different contents of ESO and ESO/(6 phr PANI/NBR) composites filled with different contents of ESO are showed in Figure S1. From Figure S1, we can find the maximum actuated strain of ESO/(4 phr PANI/NBR) composites are larger than that of ESO/(6 phr PANI/NBR) composites at the same content of ESO.



Figure S2 SEM micrographs of different composites: (a) 2 phr PANI/NBR composite,(b) 4 phr PANI/NBR composite, (c) 6 phr PANI/NBR composite, (d) 30 phr ESO/(4 phr PANI/NBR) composite, and (e) 50 phr ESO/(4 phr PANI/NBR) composite.

Figure S2 shows the typical SEM micrographs of the fractured surfaces of the NBR composites filled with different amounts of PANI and 4 phr PANI/NBR composites filled with different amounts of ESO. From Figure S2(a), Figure S2(b) and Figure S2(c), we can find that most PANI particles uniformly dispersed in the NBR matrix, ascribed to the good compatibility between the PANI particles and NBR

matrix. When polar ESO were added into the PANI/NBR composites, almost all PANI particles are dispersed in the NBR matrix individually without any contacting with each other (shown in Figure S2(d) and Figure S2(e)).



Figure S3 The (i) XPS wide-scan and (ii) N 1s core-level spectra of (a) pure NBR, (b) PANI, (c) 6 phr PANI/NBR composite, and (d) 50 phr ESO/(6 phr PANI/NBR) composite.

| Samples | Element Content (wt%) | | |
|--------------|-----------------------|-------|-------|
| | C 1s | N 1s | O 1s |
| PANI | 36.07 | 30.69 | 32.64 |
| NBR | 72.65 | 12.06 | 15.30 |
| PANI/NBR | 78.60 | 5.96 | 15.44 |
| ESO/PANI/NBR | 79.44 | 1.29 | 19.28 |

Table S1 Chemical composition of pristine PANI and NBR composites surfaces by

X-ray photoelectron spectroscopy.

Figure S3 shows the XPS wide-scan and N 1s core-level spectra of the NBR composites and PANI particles. Table S1 summarizes the chemical composition of pristine PANI and NBR composites surfaces determined by XPS. The X-ray photoelectron spectroscopy (XPS) spectra were measured using an Escalab MK II (VG Company, UK). All binding energies (BE) were calibrated by C 1s. From Figure S3 and Table S1, we can find the N1s peaks of PANI/NBR composites is similar to that of PANI, indicating the polyaniline dispersed in NBR matrix with good compatibility. After ESO were added into the PANI/NBR composite, the content of N decreased largely and content of O increased in ESO/PANI/NBR composite comparing with PANI/NBR composite.