Supplementary Information for:

Polar-nonpolar interconnected elastic networks with increased permittivity and high breakdown fields for dielectric elastomer transducers

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Figure S1: $^{29}$Si NMR and $^1$H NMR spectra of a partially cross-linked copolymer (P$_62$). The signal at -57 ppm in the $^{29}$Si spectrum is assigned to trifunctional siloxane, while the Si-H signal is seriously diminished (for comparison see Figure S5b).
**Figure S2:** Reaction monitoring with IR spectroscopy: variation of the Si-H stretching band at 2154 cm\(^{-1}\) versus Si-CH\(_3\) symmetric band at 1260 cm\(^{-1}\) due to hydrolysis of P\(_{62}\) as function of time.

**Figure S3:** Evolution of the Si-H stretching band in a cross-linked film within three weeks.
Figure S4: Image of a blend of cross-linked PDMS and P_{62} (left), P_{62}(1/3) (middle) and P_{62}^{\text{part}}(1/2) (right). Macroscopic phase separation can be seen when P_{62} is not cross-linked (the left-side photo).

Figure S5: $^1$H NMR spectrum of the extract from film P_{62}^{\text{part}}(1/2) (a), compared with the starting copolymer (b). The ratio between the Si-CH$_2$ and Si-CH$_3$ protons indicate that the extract contains little amount of un-cross-linked P_{62} and a high amount of un-cross-linked PDMS (possible macrocycles) –roughly 1:7.4. A large amount of DBDTL catalyst is, as expected, present in the extract. No signal for Si-H protons can be seen.
Figure S6: SEM image of cross-section of material P_{62}^{part}(1/2).

Figure S7: DSC scans (second heating) of P_{10} (red), PDMS (blue) and P_{10}(1/2) (black).
Figure S8: DMA curves of $P_{62}(1/2)$ in tension mode, at 1 Hz, at a heating rate of 2 °C/min.

Figure S9: The cyclic test results for materials $P_{62}(1/2)$ and $P_{10}(1/2)$. 

Table S1: Mechanical properties of the optimized materials compared to the corresponding ones with similar composition.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Young modulus (MPa)</th>
<th>Tensile strength [MPa]</th>
<th>Elongation at break [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10%</td>
<td>20%</td>
<td>50%</td>
</tr>
<tr>
<td>$P_{62}(1/3)$</td>
<td>0.75</td>
<td>0.62</td>
<td>0.41</td>
</tr>
<tr>
<td>$\text{soft}P_{62}(1/3)$</td>
<td>0.28</td>
<td>0.21</td>
<td>0.67</td>
</tr>
<tr>
<td>$P_{62}\text{part}(1/2)$</td>
<td>0.48</td>
<td>0.45</td>
<td>0.46</td>
</tr>
<tr>
<td>$\text{soft}P_{62}\text{part}(1/2)$</td>
<td>0.14</td>
<td>0.32</td>
<td>0.20</td>
</tr>
</tbody>
</table>

**Figure S10:** Calculated and experimental actuation strain at 30 V/µm (left) and at breakdown (right).

**Figure S11:** Stress-strain curve (left) and electromechanical response (right) of all-polymer composites compared with the interconnected network with similar composition.