Supporting Information

**Significantly improved electromechanical performance of dielectric elastomers via alkyl side-chain engineering**

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1. GPC curves of ATPDEs
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3. TEM images of ATPDEs
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6. DMA data of ATPDEs
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1. GPC curves of ATPDEs

Fig. S1 GPC curves of chain extensions during the synthesis of ATPDEs. a) SEAS, b) SBAS) and c) SEHAS. (The black curves represent polystyrene in the end of the first stage of the polymerization, the red curves represent polystyrene-b-poly(alkylacrylate) diblock copolymer in the end of the second stage of the polymerization, the blue curves represent polystyrene-b-poly(alkylacrylate)-b-polystyrene triblock copolymer.)

2. 1H NMR spectra of ATPDEs

Fig. S2 1H NMR spectra of ATPDEs. a) SEAS, b) SBAS) and c) SEHAS. (The signals from hydrogen of phenyl groups and the signals from hydrogen of methyl groups was used to calculate the composition of the copolymers.)
3. TEM images of ATPDEs

(a)  
(b)  
(c)  

Fig. S3 TEM images of ATPDEs. a) SEAS, b) SBAS) and c) SEHAS. (The scale bar is 100nm)

4. SAXS profiles of ATPDEs

The d calculated by $d=\frac{2\pi}{q}$ is calculated, which illustrate the size of PS domains is around 53 nm for all ATPDEs corresponding to the AFM and TEM analysis. However, we could not distinguish the morphology of ATPDEs accurately due to the weak phase separation in these triblock system.

![SAXS profiles of ATPDEs. a) SEAS, b) SBAS) and c) SEHAS.](image)

5. DSC curves of ATPDEs
6. DMA data of ATPDEs

![DMA data](image)

Fig. S6 DMA data of ATPDEs. a) Storage modulus and b) loss factor as a function of temperature.

7. Dielectric properties of ATPDEs
Fig. S7 Dielectric properties of ATPDEs. a) relative permittivity, b) imaginary permittivity, c) loss factor, and d) conductivity of ATPDEs as a function of frequency at 25 °C.

8. Havriliak–Negami relaxation of ATPDEs

Havriliak–Negami relaxation could be indicated as:

\[ \varepsilon'(\omega) = \varepsilon_\infty + \frac{\Delta \varepsilon}{1 + i\omega\tau^\alpha}^{\beta} \]

where \( \varepsilon_\infty \) is the permittivity at the high frequency limit, \( \Delta \varepsilon = \varepsilon_s - \varepsilon_\infty \) where \( \varepsilon_s \) is the static, low frequency permittivity, and \( \tau \) is the dielectric relaxation time. The exponents \( \alpha \) and \( \beta \) describe the asymmetry and broadness of the corresponding spectra.

Table S1 Parameter of Havriliak–Negami relaxation of ATPDEs

<table>
<thead>
<tr>
<th>Parameter</th>
<th>SEAS</th>
<th>SBAS</th>
<th>SEHAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \varepsilon_\infty )</td>
<td>5.01</td>
<td>5.02</td>
<td>5.03</td>
</tr>
<tr>
<td>( \Delta \varepsilon )</td>
<td>0.1</td>
<td>0.12</td>
<td>0.11</td>
</tr>
<tr>
<td>( \alpha )</td>
<td>0.5</td>
<td>0.55</td>
<td>0.52</td>
</tr>
<tr>
<td>( \beta )</td>
<td>0.3</td>
<td>0.35</td>
<td>0.33</td>
</tr>
</tbody>
</table>

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9. Brief illustration of electromechanical performance test

The electromechanical performance without prestretch was conducted by a diaphragm actuator (diameter 30mm) with a bias air pressure of 100 Pa. The low pressure was not enough to prestretch the film before actuation. When powered on, the ATPDE films plump up to a dome shape with a radius $r$ and a raised height $h$. The area strain ($S_{area}$) can be calculated by\(^1\):

$$S_{area} = \frac{(h^2 + r^2) - (h_0^2 + r_0^2)}{h_0^2 + r_0^2}$$

**Reference:**