Supporting Information

Synthesis of solvent-free processable and on-demand cross-linkable dielectric elastomers for actuators

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Figure S1 <sup>1</sup>H NMR spectrum of P1 ( $M_n$  = 2500 g/mol) (top) and ( $M_n$  = 5600 g/mol) bottom in CDCl<sub>3</sub>.





Figure S4 <sup>13</sup>C NMR spectrum of **P3** ( $M_n$  = 3500 g/mol) in CDCl<sub>3</sub>.



Figure S6 GPC elution curves of **P3.2**. *M*<sub>n</sub>=3000, *M*<sub>w</sub>= 18000, PDI= 5.5



Figure S7 GPC elution curves of **P4**.  $M_n$ = 150.000,  $M_w$ = 450.000, PDI= 2.4.



Figure S8 GPC elution curves of the depolymerization of P4 under basic conditions.



Figure S9 GPC elution curves of **P5**.  $M_n$  = 8500,  $M_w$  = 20000, PDI = 2.4.





Figure S12 GPC elution curves of R(SH)<sub>3</sub>.



Figure S13 Viscosity of P3.2 measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S14 Viscosity of P3 measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S15 Viscosity of P3-1 measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S16 Viscosity of P3-2 measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S17 Viscosity of **P3-3** measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S18 Viscosity of **P3-4** measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S19 Viscosity of  $\alpha'\omega$ -OH-PDMS ( $M_n$  = 28 kDa) measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S20 Viscosity of  $\alpha'\omega$ -OH-PDMS ( $M_n$  = 63 kDa) measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S21 Viscosity of  $\alpha'\omega$ -OH-PDMS ( $M_n$  = 139 kDa) measured in the frequency range of 1-10 Hz. Five measurements were performed.



Figure S22 TGA curve of P3 in air.



Figure S23 TGA curve of **R(SH)<sub>3</sub>** in air.



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Figure S25 TGA curve of AB113729 in air.



Figure S26 DMA of E3-1. Four samples were measured.



Figure S27 DMA of **E3-2**. Three samples were measured.



Figure S28 DMA of E3-3. Three samples were measured.



Figure S29 DMA of E3-4. Three samples were measured.



Figure S30 DMA of  $\ensuremath{\text{E5}}$  . Three samples were measured.



Figure S31 DMA of Elastosil®Film. Three samples were measured.



Figure S32 DMA of **E5** measured 24h/48h/72h after synthesis. The average curve of 3 measurements is given. The identical sample was measured.



Figure S33 Tensile tests of **E3-1**. Three independent tests were performed.



Figure S34 Tensile tests of **E3-2**. Three independent tests were performed.



Figure S35 Tensile tests of **E3-3**. Three independent tests were performed.



Figure S36 Tensile tests of E3-4. Three independent tests were performed.



Figure S37 Tensile tests of E5. Three independent tests were performed.



Figure S38 Tensile tests of Elastosil®Film. Three independent tests were performed.



Figure S39 DSC curves of E3-1.









Figure S42 DSC curves of E3-4.



Figure S43 DSC curves of E5.



Figure S44 TGA curve of **E3-1** in air.



Figure S45 TGA curve of E3-2 in air.



Figure S46 TGA curve of **E3-3** in air.



Figure S47 TGA curve of E3-4 in air.



Figure S48 TGA curve of E5 in air.



Figure S49 Dielectric properties of P3 and R(SH)<sub>3</sub>.



Figure S50 Lateral strain in x-and y-direction (strain 1 and strain 2) of E3-1 as a function of the applied electric field.



Figure S51 Lateral strain in x-and y-direction (strain 1 and strain 2) of E3-2 as a function of the applied electric field.



Figure S52 Lateral strain in x-and y-direction (strain 1 and strain 2) of E3-3 as a function of the applied electric field.



Figure S53 Lateral strain in x-and y-direction (strain 1 and strain 2) of E3-4 as a function of the applied electric field.



Figure S54 Lateral strain in x-and y-direction (strain 1 and strain 2) of E3-4 as a function of the applied electric field.



Figure S55 Lateral strain in x-and y-direction (strain 1 and strain 2) of **Elastosil®Film** as a function of the applied electric field.



Figure S56 Average lateral strain of **E3-Y** and **Elastosil®Film** as a function of the applied electric field (DC) operated at 1 Hz. The electric field was 80 V/µm for **Elastosil®Film**, 30 V/µm for **E3-1**, 25 V/µm for **E3-2/E3-3**, and 20 Vµm for **E3-4**.



Figure S57 Average lateral strain of **E3-Y** and **Elastosil®Film** as a function of the applied electric field (DC) operated at 2 Hz. The electric field was 80 V/µm for **Elastosil®Film**, 30 V/µm for **E3-1**, 25 V/µm for **E3-2/E3-3**, and 20 Vµm for **E3-4**.



Figure S58 Average lateral strain of E3-Y and Elastosil®Film as a function of the applied electric field (DC) operated at 4 Hz. The electric field was 80 V/µm for Elastosil®Film, 30 V/µm for E3-1, 25 V/µm for E3-2/E3-3, and 20 Vµm for E3-4.



time (s) Figure S59 Average lateral strain of E3-Y and Elastosil®Film as a function of the applied electric field (DC) operated at 8 Hz. The electric field was 80 V/μm for Elastosil®Film, 30 V/μm for E3-1, 25 V/μm for E3-2/E3-3, and 20 Vμm for E3-4.



Figure S60 10 x 10.000 operation cycles at 8 Hz at 25 V/ $\mu$ m of a circular DEA test device constructed from a 75  $\mu$ m-thin film of **E3-1** with an area diameter of 8 mm. Strain 1 is defined as the lateral strain in x-direction. Strain 2 is defined as the lateral strain in y-direction.



Figure S61 5 x 10.000 operation cycles + 1 x 20.0000 at 8 Hz at 29 V/ $\mu$ m of a circular DEA test device constructed from a 85  $\mu$ m-thin film of **E3-2** with an area diameter of 8 mm. Strain 1 is defined as the lateral strain in x-direction. Strain 2 is defined as the lateral strain in y-direction.



Figure S62 3 x 10.000 + 2 x 30.0000 DEA operation cycles at 8 Hz at 25 V/ $\mu$ m of a circular DEA test device constructed from a 90  $\mu$ m-thin film of **E3-3** with an area diameter of 8 mm. Strain 1 is defined as the lateral strain in x-direction. Strain 2 is defined as the lateral strain in y-direction.



Figure S63 1 x 10.000 DEA operation cycles at 8 Hz at 15 V/ $\mu$ m of a circular DEA test device constructed from a 100  $\mu$ m-thin film of **E3-4** with an area diameter of 8 mm. Strain 1 is defined as the lateral strain in x-direction. Strain 2 is defined as the lateral strain in y-direction.



Figure S64 2 x 50.000 and 2 x 40.000 DEA operation cycles at 8 Hz at 25 V/ $\mu$ m of a circular DEA test device constructed from a 85  $\mu$ m-thin film of E5 with an area diameter of 8 mm. Strain 1 is defined as the lateral strain in x-direction. Strain 2 is defined as the lateral strain in y-direction.