

## Electronic supplementary information (ESI)

### Elastomers with tunable dielectric and electromechanical properties

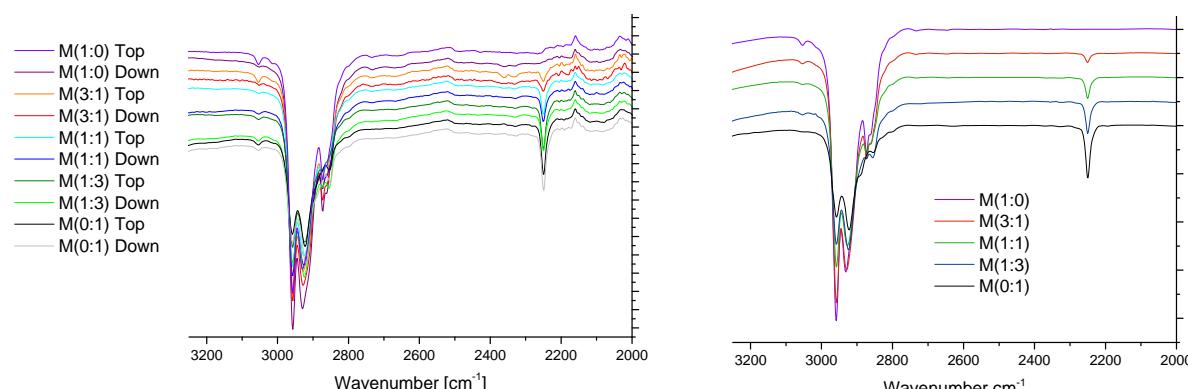
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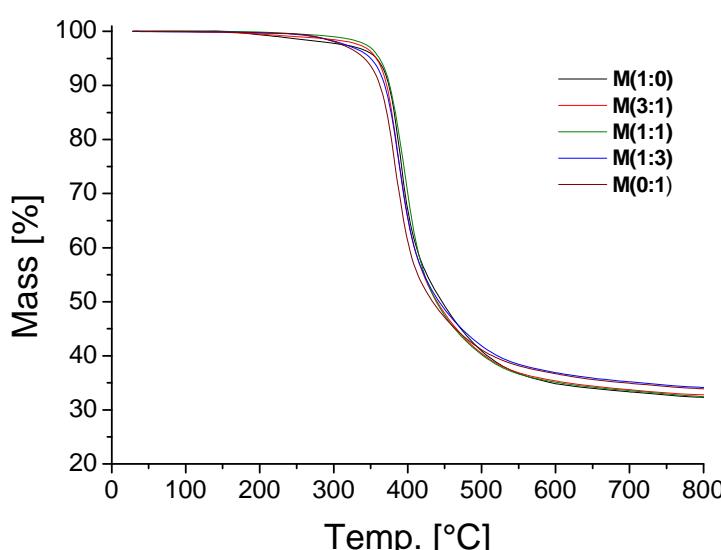
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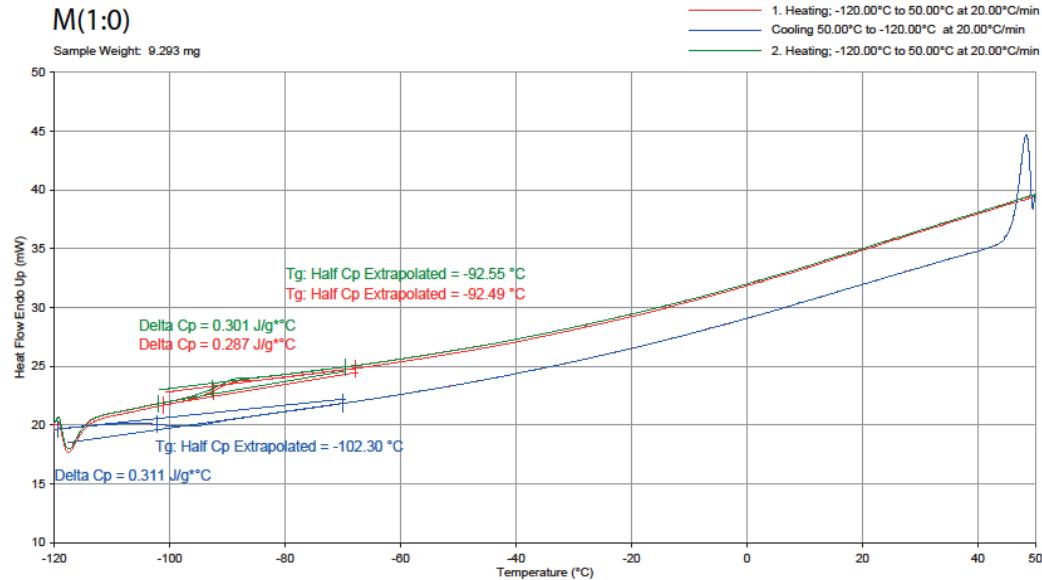
**Figure S1.** IR spectra of the two film surfaces of different materials (top layer exposed to the glass plate and down layer exposed to the Teflon substrate) (left) and of the ground films in KBr (right).



**Figure S2.** TGA curves of  $\mathbf{M}(x:y)$  conducted at a heating rate of  $20\text{ }^{\circ}\text{C min}^{-1}$  under nitrogen.

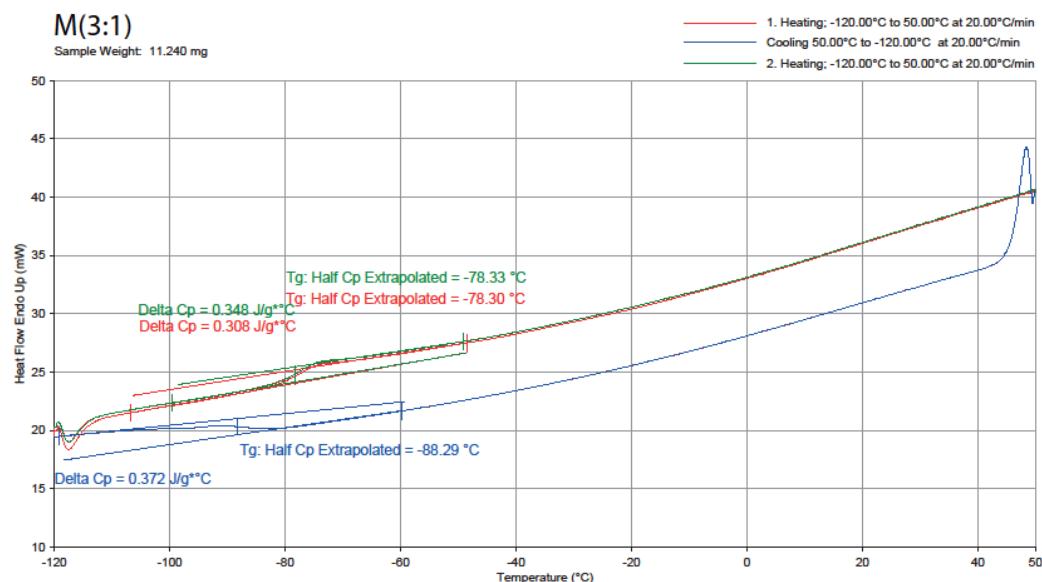
### M(1:0)

Sample Weight: 9.293 mg



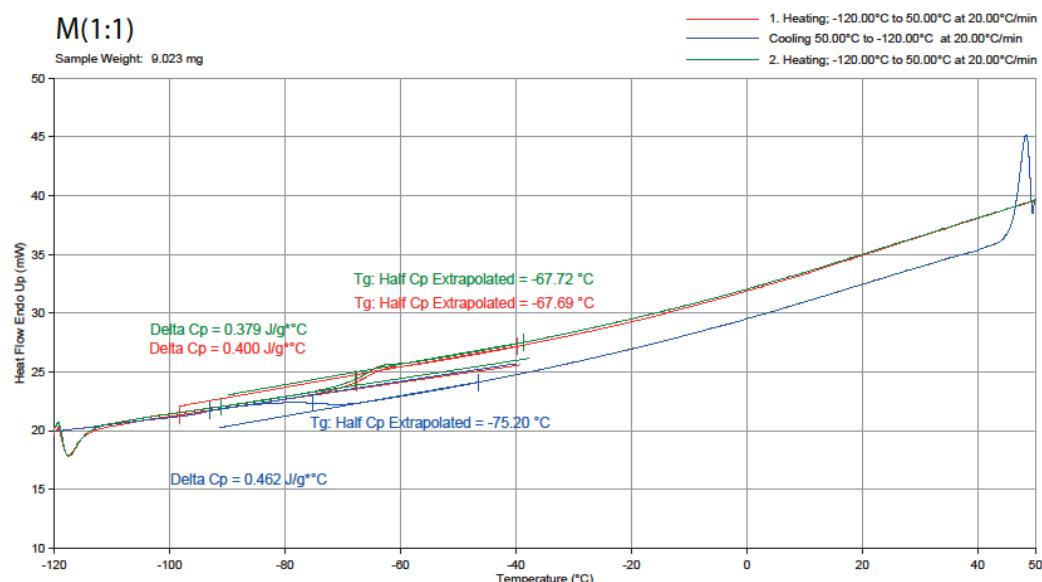
### M(3:1)

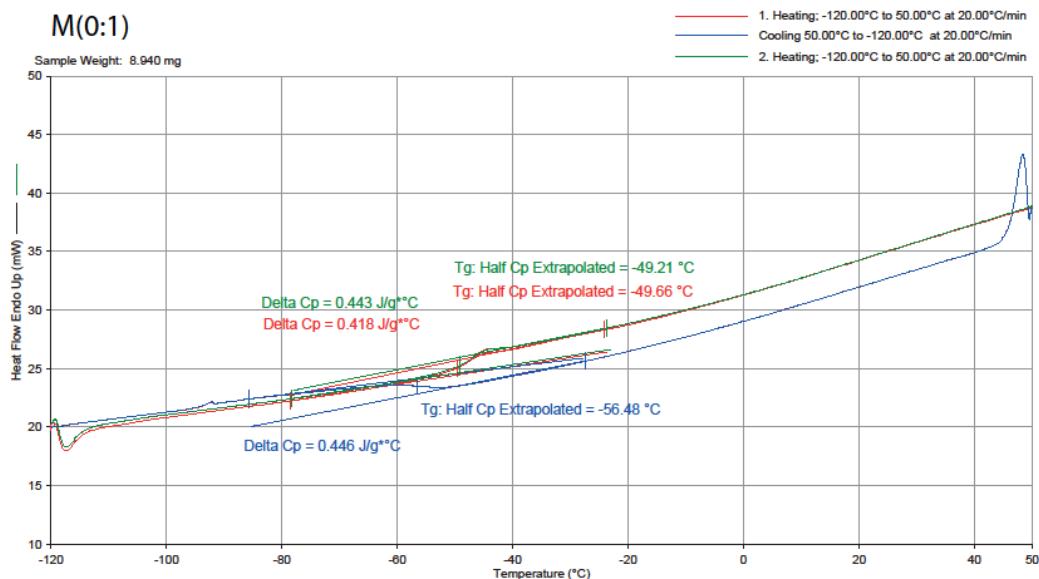
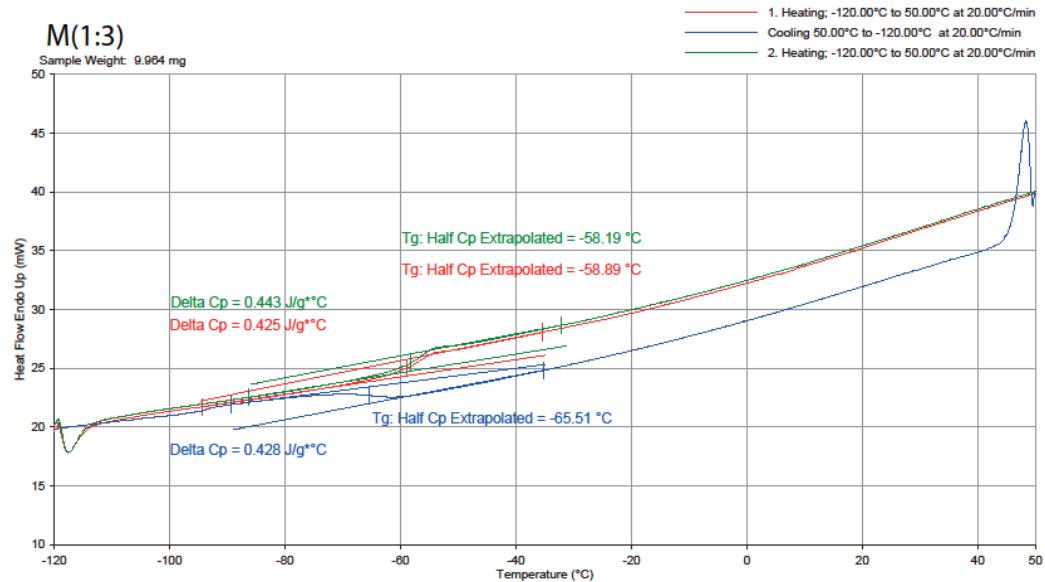
Sample Weight: 11.240 mg



### M(1:1)

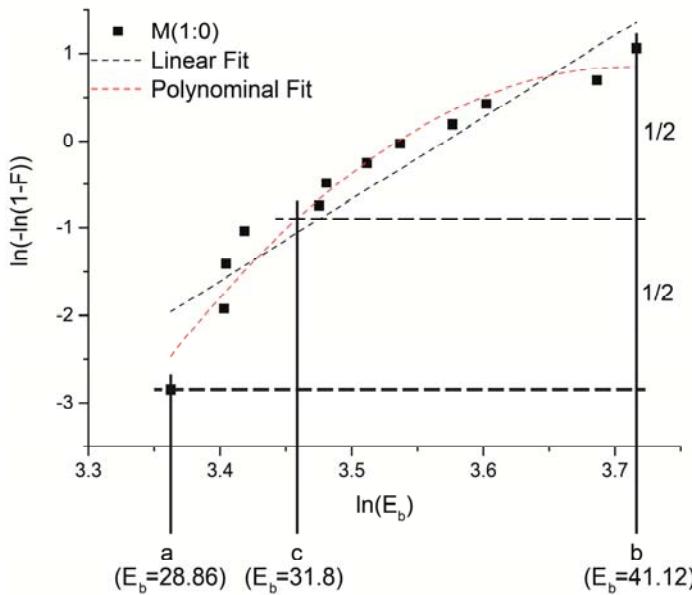
Sample Weight: 9.023 mg





**Figure S3.** DSC curves of different materials **M(x:y)**. An increase in  $T_g$  with CN content can be seen.

Calculation of the Weibull parameter for **M(1:0)** when the Weibull location parameter  $\gamma$  is not neglected. From the Weibull plot the two extreme points on the y axis with the corresponding  $E_b$  values (a and b) are taken. The physical distance on the y axis was divided by two and the corresponding  $E_b$  value (c) was found. (Figure S4).



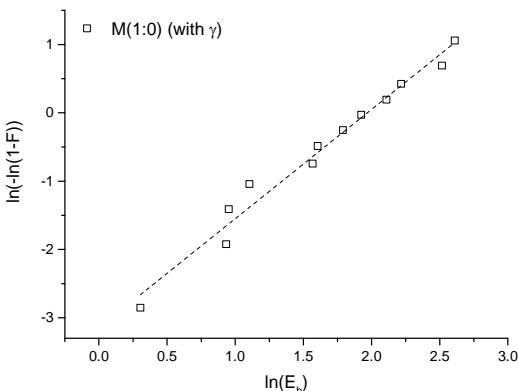
**Figure S4.** determination of a, b and c values required to estimate  $\gamma$ .

$\gamma$  was calculated according to equation (1):

$$\gamma = c - \frac{(b-c)(c-a)}{(b-c)-(c-a)} \quad (1)$$

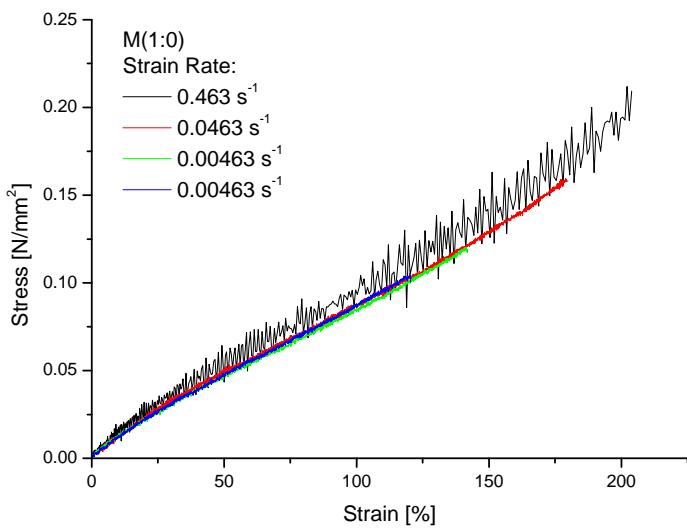
and found to be 27.4.

This value was included in the calculation for the Weibull plot from which  $\beta$  and  $\eta_p$  were obtained as previously described in the paper (Figure S5). The location parameter  $\gamma$  has to be added to  $\eta_p$  to obtain the scale parameter  $\eta$

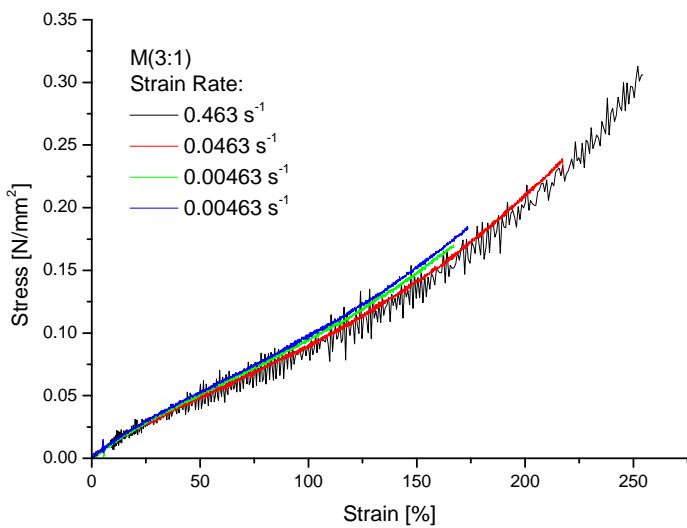


**Figure S5.** Weibull plot for M(1:0) considering  $\gamma$ . Dotted line represents the linear fit.

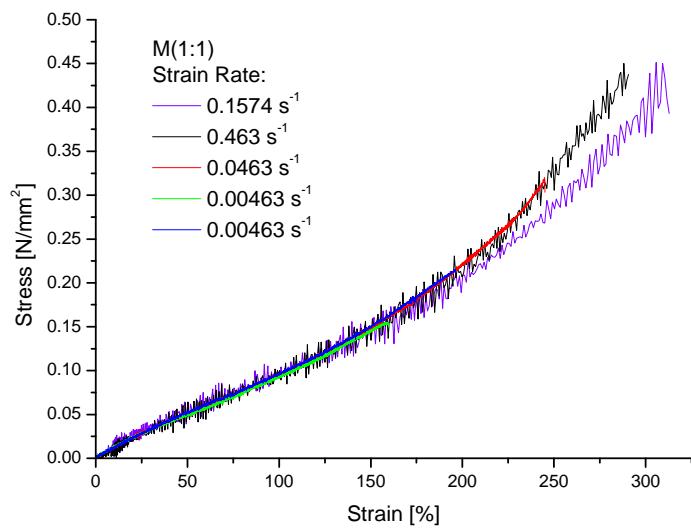
Sample	Dielectric breakdown	$E_b$ Stab	Weibull $\beta$ -parameter	Weibull $\eta$ -parameter	Weibull $\gamma$ -parameter	Weibull plot Intercept	$R^2$ of linear fit
M(1:0)	33.8	4.3	1.6	34.7	27.5	-3.15	0.97



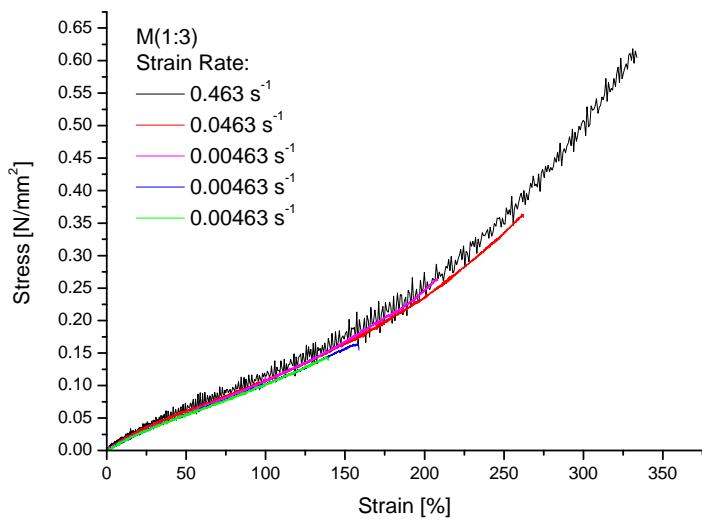
**Figure S6.** Stress-strain curves for material **M(1:0)** from standard tensile tests at various initial strain rates ( $0.463\text{ s}^{-1}$ ,  $0.0463\text{ s}^{-1}$ ,  $0.00463\text{ s}^{-1}$ , and  $0.00463\text{ s}^{-1}$ ).



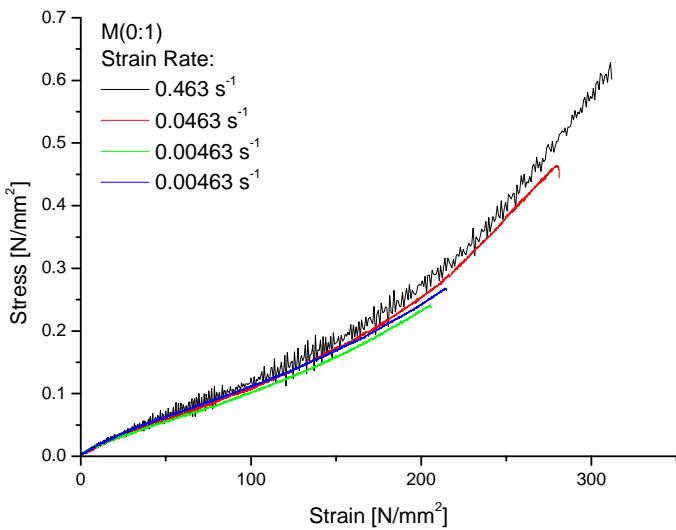
**Figure S7.** Stress-strain curves for material **M(3:1)** from standard tensile tests at various initial strain rates ( $0.463\text{ s}^{-1}$ ,  $0.0463\text{ s}^{-1}$ ,  $0.00463\text{ s}^{-1}$ , and  $0.00463\text{ s}^{-1}$ ).



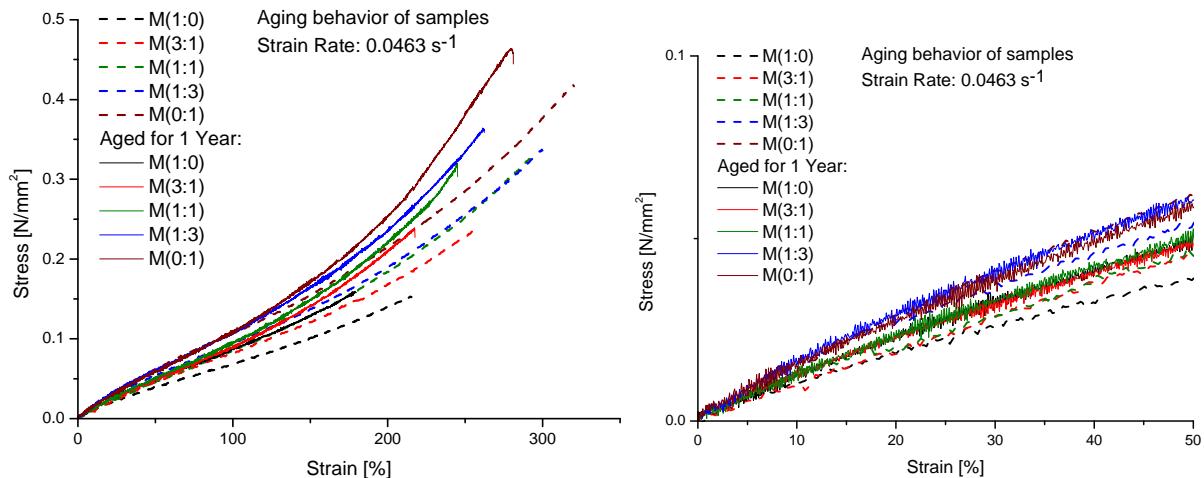
**Figure S8.** Stress-strain curves for material **M(1:1)** from standard tensile tests at various initial strain rates ( $0.463\text{ s}^{-1}$ ,  $0.0463\text{ s}^{-1}$ ,  $0.00463\text{ s}^{-1}$ , and  $0.00463\text{ s}^{-1}$ ).



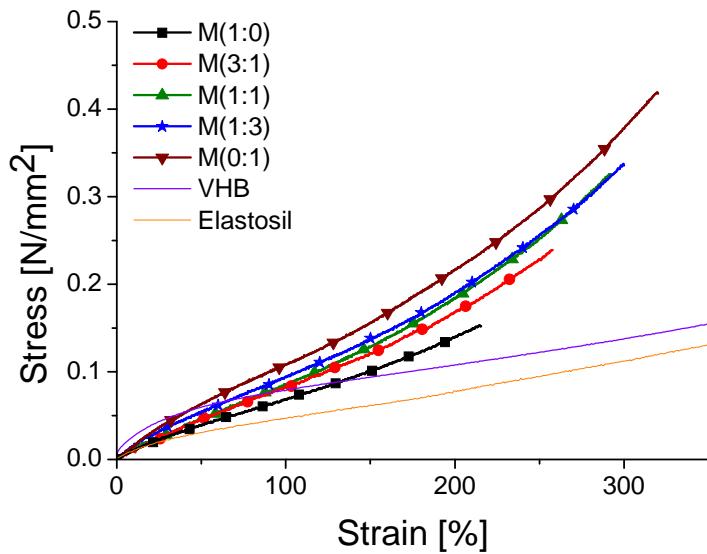
**Figure S9.** Stress-strain curves for material **M(1:3)** from standard tensile tests at various initial strain rates ( $0.463\text{ s}^{-1}$ ,  $0.0463\text{ s}^{-1}$ ,  $0.00463\text{ s}^{-1}$ , and  $0.00463\text{ s}^{-1}$ ).



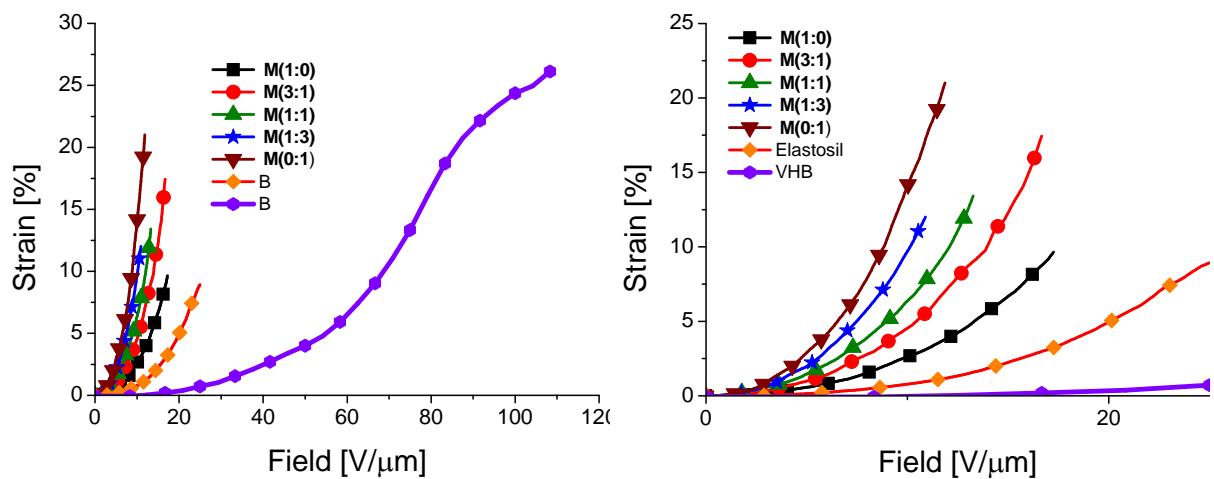
**Figure S10.** Stress-strain curves for material **M(1:0)** from standard tensile tests at various initial strain rates ( $0.463\text{ s}^{-1}$ ,  $0.0463\text{ s}^{-1}$ ,  $0.00463\text{ s}^{-1}$ , and  $0.00463\text{ s}^{-1}$ ).



**Figure S11.** Stress-strain curves for materials **M(x:y)** from standard tensile tests at initial strain rate of  $0.0463\text{ s}^{-1}$  after the synthesis and after 1 year. A slight increase in the stiffness after one year aging can be seen.



**Figure S12.** Stress-strain curves for materials  $\mathbf{M}(x:y)$ , VHB and Elastosil from standard tensile tests at  $50 \text{ mm min}^{-1}$ . The stress-strain curves were averaged from three independent tests.



**Figure S13.** Lateral actuation strain as function of electric field for materials  $\mathbf{M}(x:y)$ , Elastosil and VHB which were 28.6% biaxial prestrained except VHB for which 300% bilateral strain was used.