Deformation behavior of crosslinked polyurea elastomers obtained via sol-gel chemistry: experimental determination and constitutive modelling

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Appendix A

Heuristic function used for fitting uniaxial test experiments

The equation that describes the increase of true stress as a function of the applied true strain is given by

$$\sigma_t(\varepsilon_t) = a\varepsilon_t + b(e^{c\varepsilon_t} - 1) + d(1 - e^{-f\varepsilon_t}) \qquad \text{Equation (SI-1)}$$

The corresponding fitting parameter "a" corresponds to the linear behavior of the elastic modulus, "b" and "c" are the fitting parameters of the positive exponential growth, and "d" and "f" are the fitting parameters of the negative exponential decay.

The elastic modulus is calculated from the derivative of Equation SI-1 giving the following equation:

$$E(\varepsilon_t) = \frac{d\sigma_t}{d\varepsilon_t} = a + bce^{c\varepsilon_t} + dfe^{-f\varepsilon_t} \qquad \text{Equation (SI-2)}$$

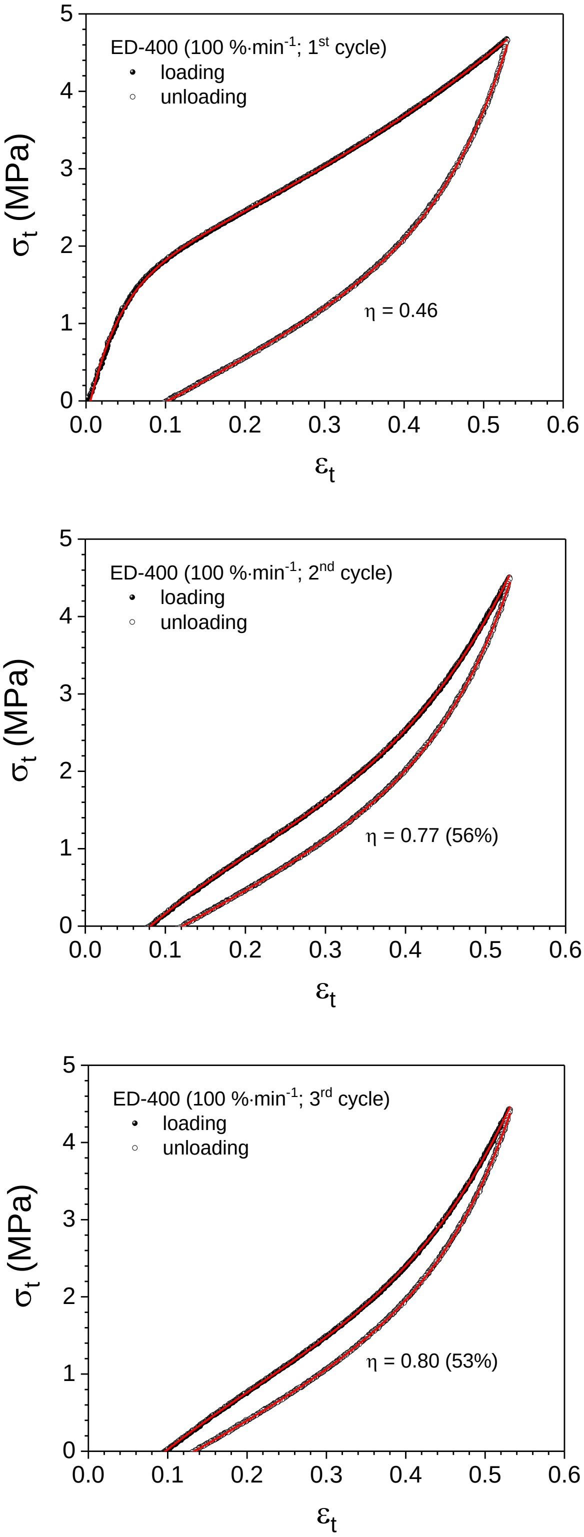


Figure SI-1. Uniaxial tensile test experiments for the PU elastomers ED-400 during three loading-unloading cycles at the strain rate of 100 % min^{-1} with the corresponding fitting curves (red)

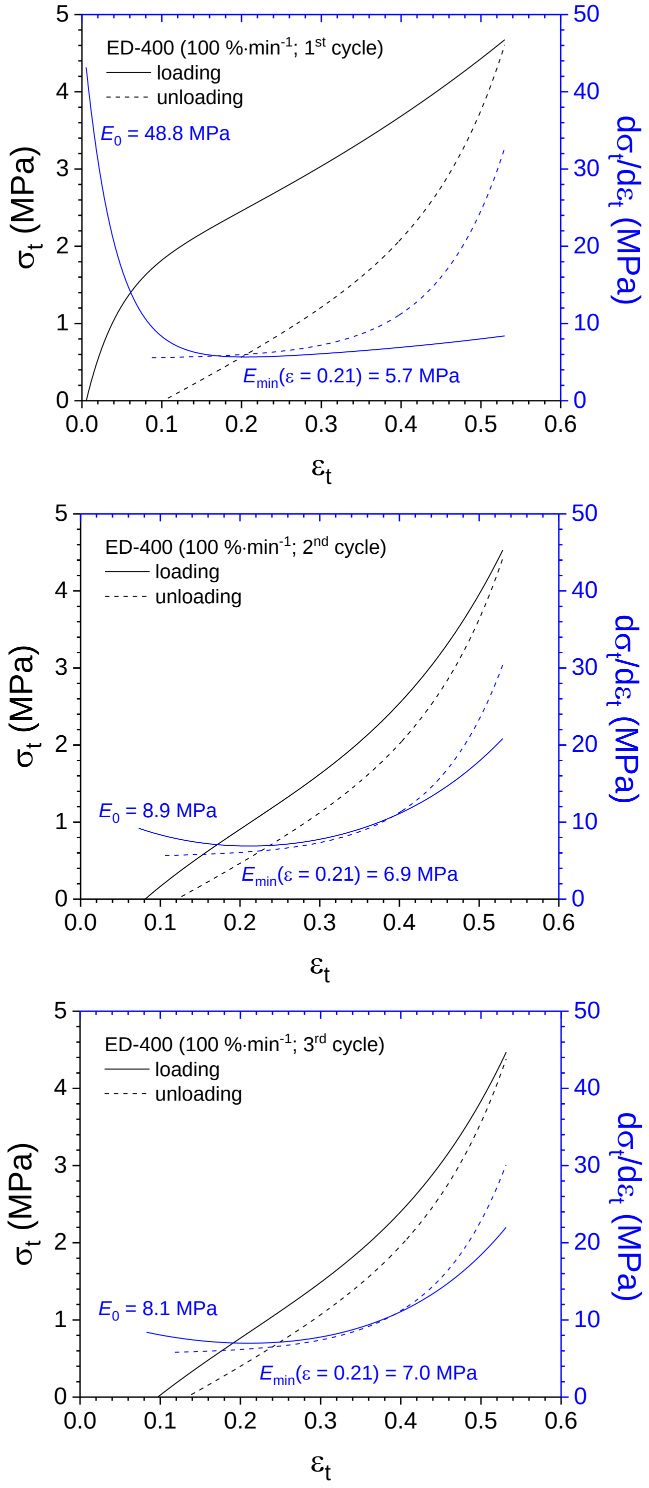


Figure SI-2. Fitting curves (black) and the corresponding derivatives (blue) for the three loading-unloading cycles at the strain rate of 100 % min^{-1} for the PU elastomers ED-400.

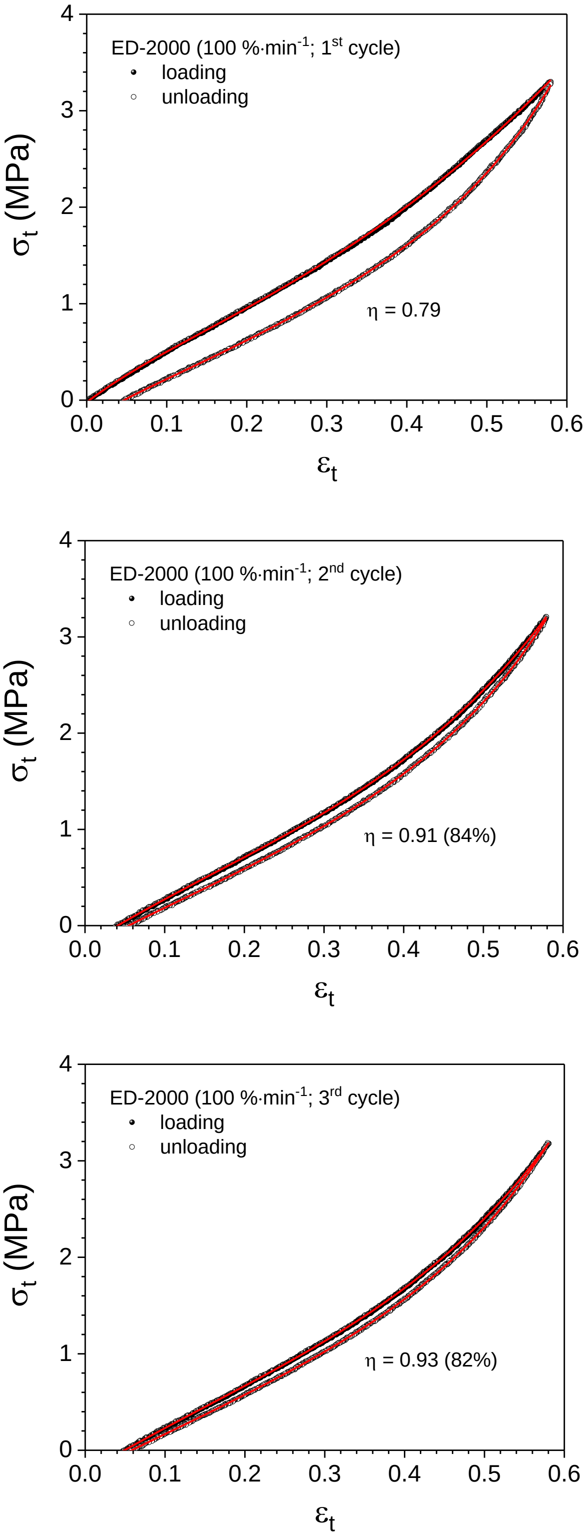


Figure SI-3. Uniaxial tensile test experiments for the PU elastomers ED-2000 during three loading-unloading cycles at the strain rate of 100 % min^{-1} with the corresponding fitting curves (red)

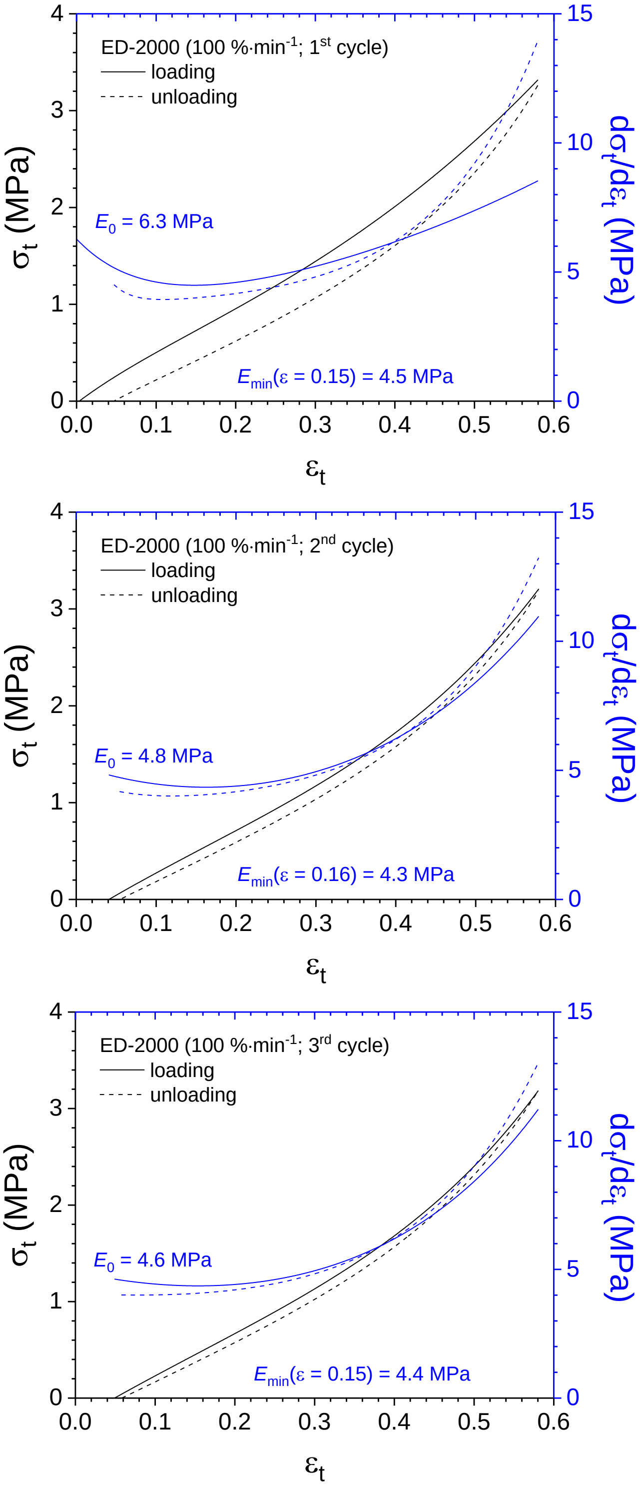


Figure SI-4. Fitting curves (black) and the corresponding derivatives (blue) for the three loading-unloading cycles at the strain rate of 100 % min^{-1} for the PU elastomers ED-2000.

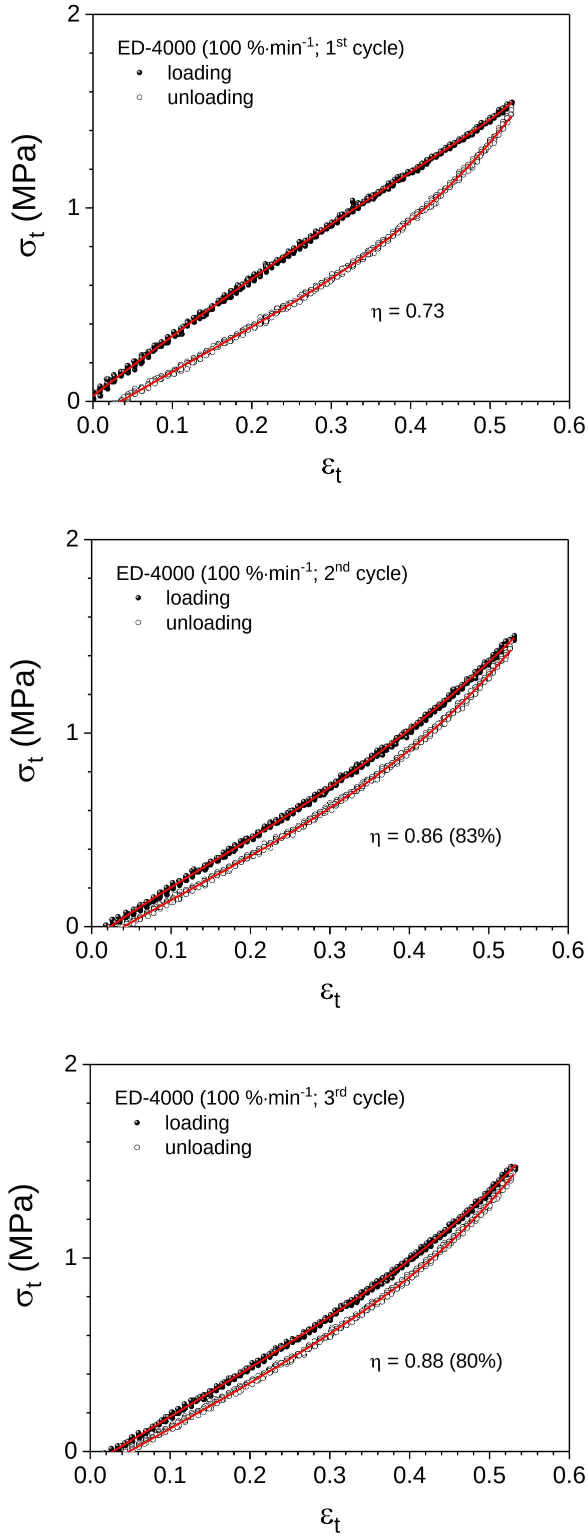


Figure SI-5. Uniaxial tensile test experiments for the PU elastomers ED-4000 during three loading-unloading cycles at the strain rate of 100 % min^{-1} with the corresponding fitting curves (red)

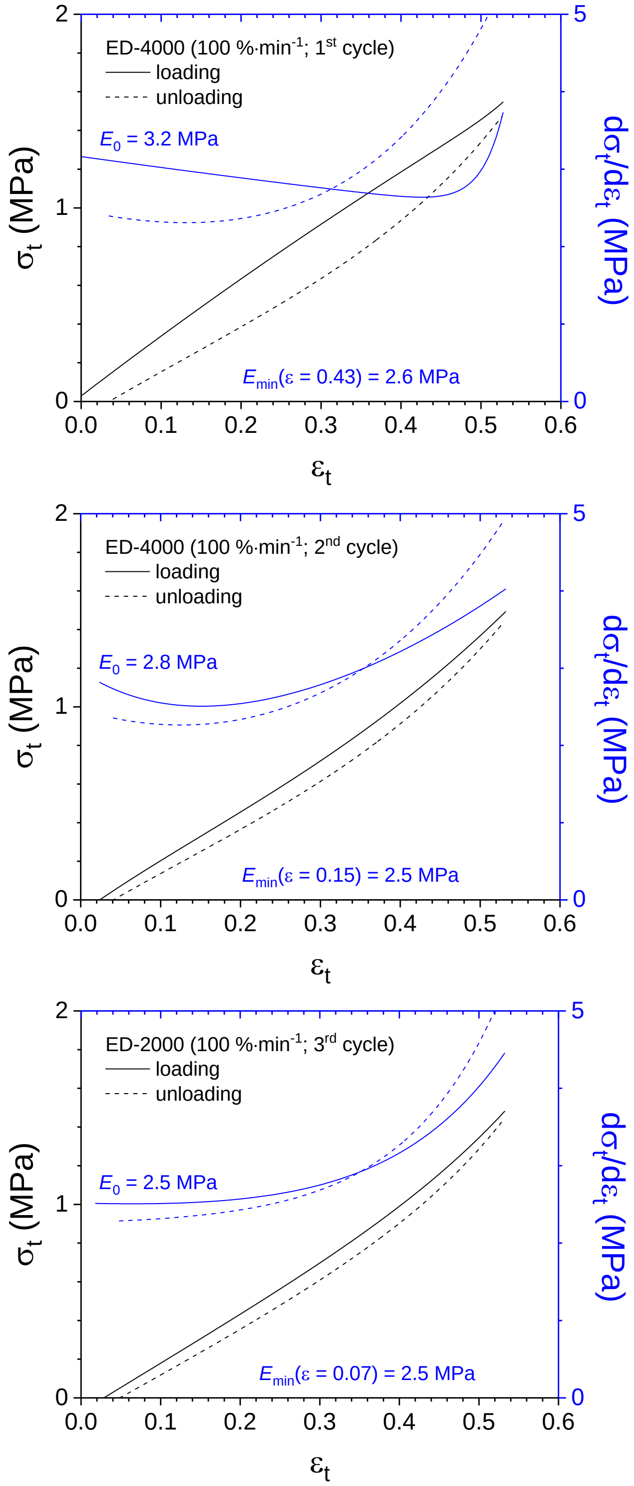


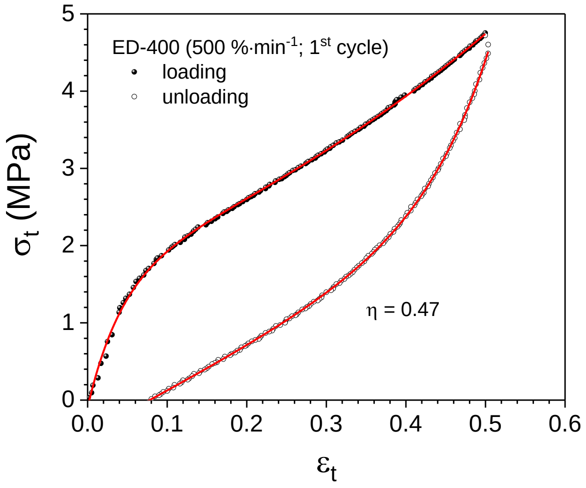
Figure SI-6. Fitting curves (black) and the corresponding derivatives (blue) for the three loading-unloading cycles at the strain rate of 100 % $min^{-1}$for the PU elastomers ED-4000.

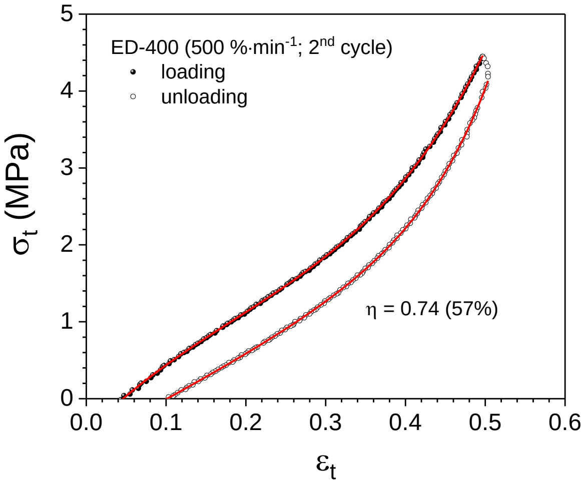
Table SI-1. Fitting parameters obtained from the uniaxial tensile test experiments for the three PU elastomers ED-400, ED-2000 and ED-4000 during the three loading-unloading cycles at the strain rate of 100 % min^{-1}.

Sample	**Cycle #**	**a**	**b**	**c**	**d**	**f**
ED-400	1st loading	3.0	0.58	2.5	1.7	26
	unloading	5.4	0.0042	12	-	-
	2nd loading	-	0.21	5.5	2.0	5.9
	unloading	5.5	0.0057	11	-	-
	3rd loading	-	0.12	6.3	2.4	4.2
	unloading	5.6	0.0056	11	-	-
ED-2000	1st loading	-	1.6	1.8	0.29	11
	unloading	3.6	0.015	7.7	0.13	44
	2nd loading	-	0.31	3.8	1.1	3.8
	unloading	3.5	0.022	7.2	0.069	16
	3rd loading	-	0.21	4.3	1.5	2.7
	unloading	1.2	0.047	6.2	2.1	1.3
ED-4000	1st loading	-	-	-	6.9	0.46
	unloading	0.20	0.053	5.2	1.1	1.9
	2nd loading	-	0.78	1.9	0.23	6.5
	unloading	-	0.18	3.7	0.59	3.1
	3rd loading	0.45	0.011	6.6	5.4	0.37
	unloading	-	-	-	34.00	0.0066

Table SI-2. Initial elastic modulus (E_0), minimum elastic modulus (E_{min}), hysteresis (η) and energy recovery (ΔU) obtained from the uniaxial tensile test experiments for the three PU elastomers ED-400, ED-2000 and ED-4000 during the three loading-unloading cycles at the strain rate of 100 % min^{-1}.

Sample	**Cycle #**	**E_0 (MPa)**	**E_{min} (MPa)**	**η**	**ΔU**
ED-400	1st	48.8	5.7	0.46	
	2nd	8.9	6.9	0.77	56%
	3rd	8.1	7.0	0.80	53%
ED-2000	1st	6.3	4.5	0.79	
	2nd	4.8	4.3	0.91	84%
	3rd	4.6	4.4	0.93	82%
ED-4000	1st	3.2	2.6	0.73	
	2nd	2.8	2.5	0.86	83%
	3rd	2.5	2.5	0.88	80%





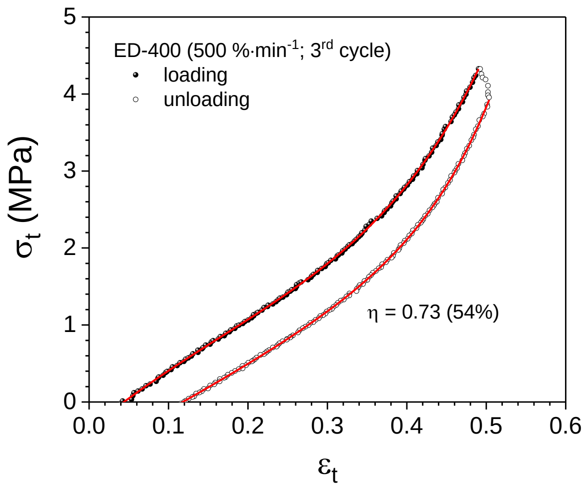


Figure SI-7. Uniaxial tensile test experiments for the PU elastomers ED-400 during three loading-unloading cycles at the strain rate of 500 % min^{-1} with the corresponding fitting curves (red)

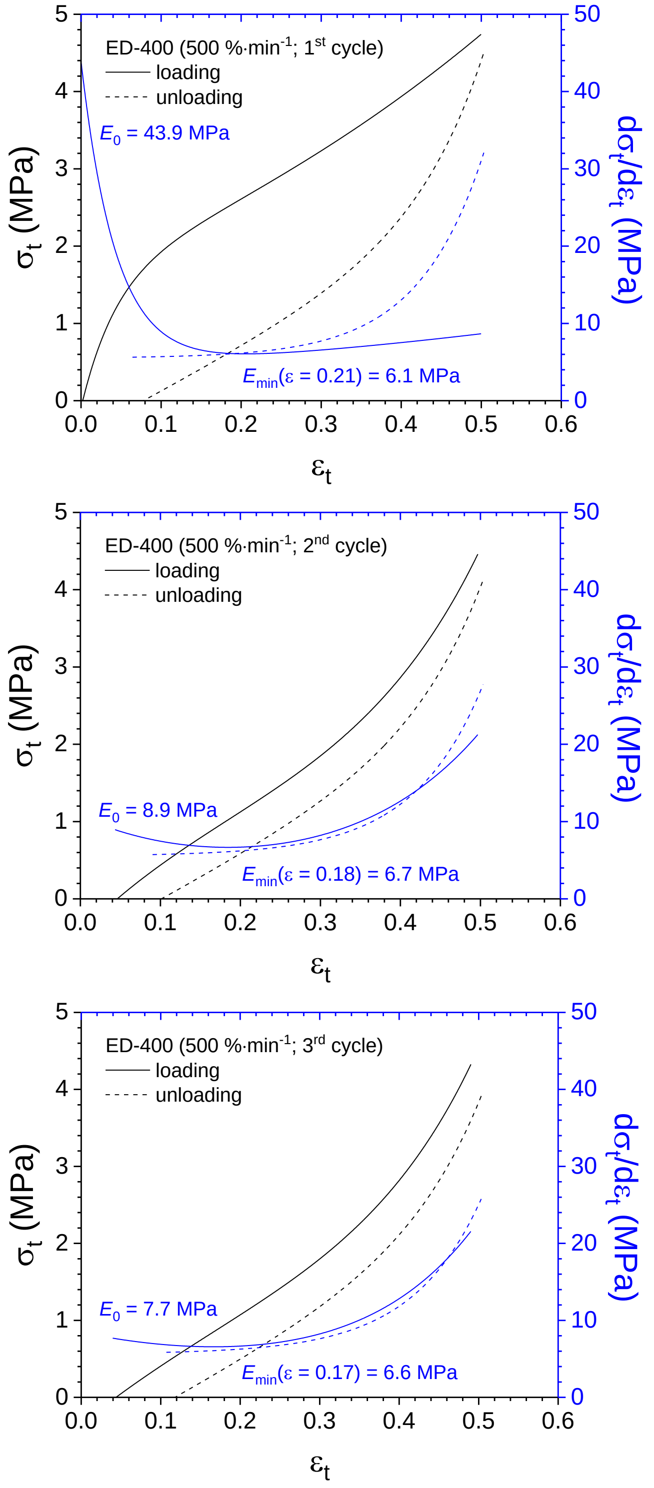


Figure SI-8. Fitting curves (black) and the corresponding derivatives (blue) for the three loading-unloading cycles at the strain rate of 500 % $min^{-1}$for the PU elastomers ED-400.

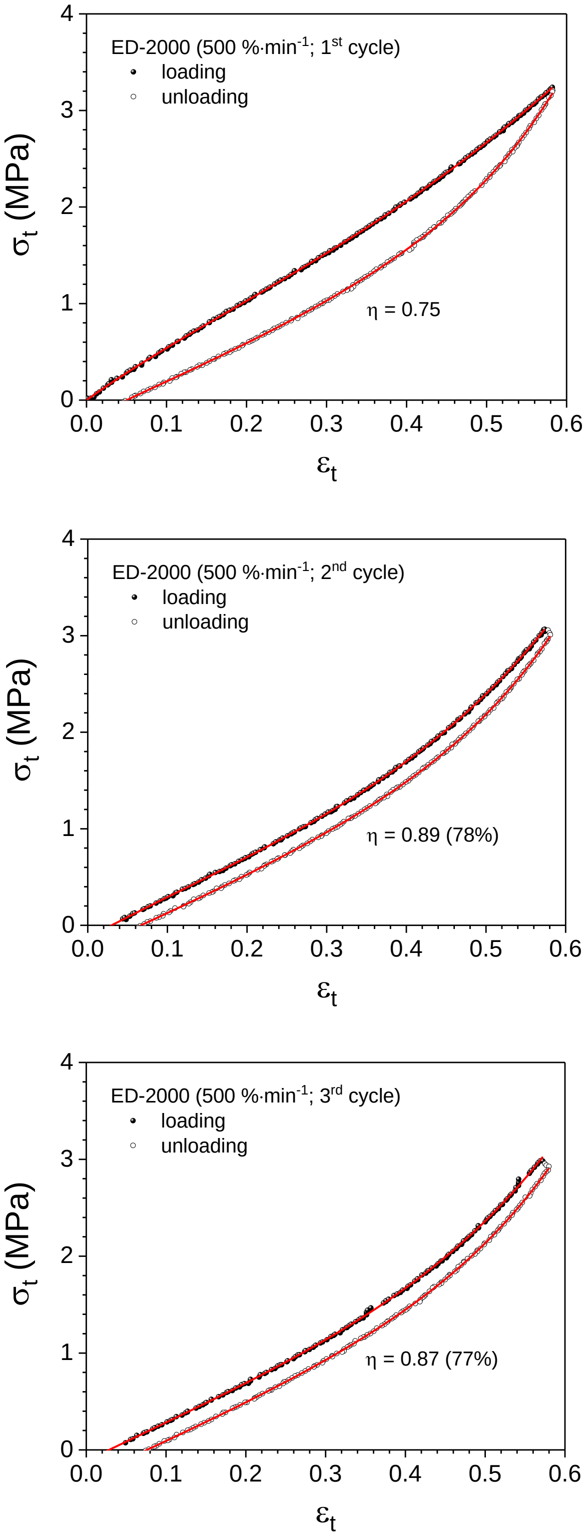


Figure SI-9. Uniaxial tensile test experiments for the PU elastomers ED-2000 during three loading-unloading cycles at the strain rate of 500 % min^{-1} with the corresponding fitting curves (red)

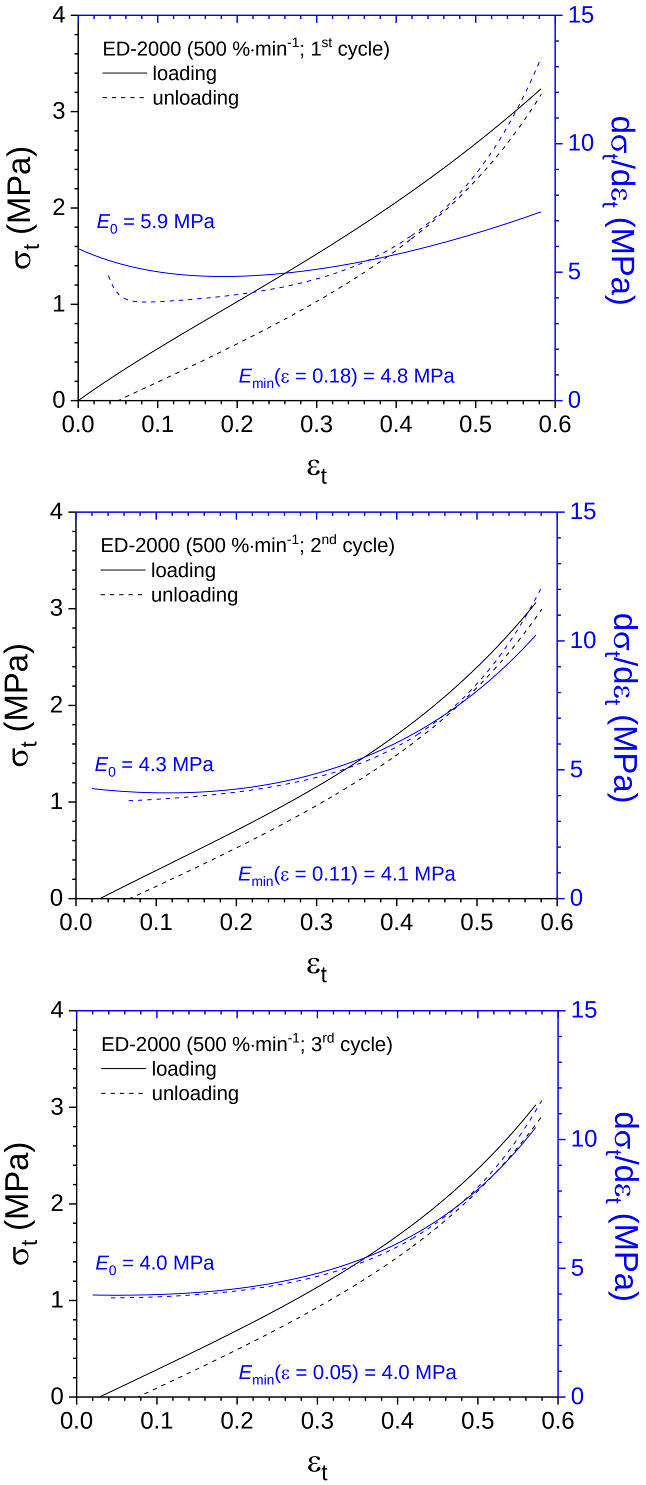


Figure SI-10. Fitting curves (black) and the corresponding derivatives (blue) for the three loading-unloading cycles at the strain rate of 500 % min^{-1} for the PU elastomers ED-2000.

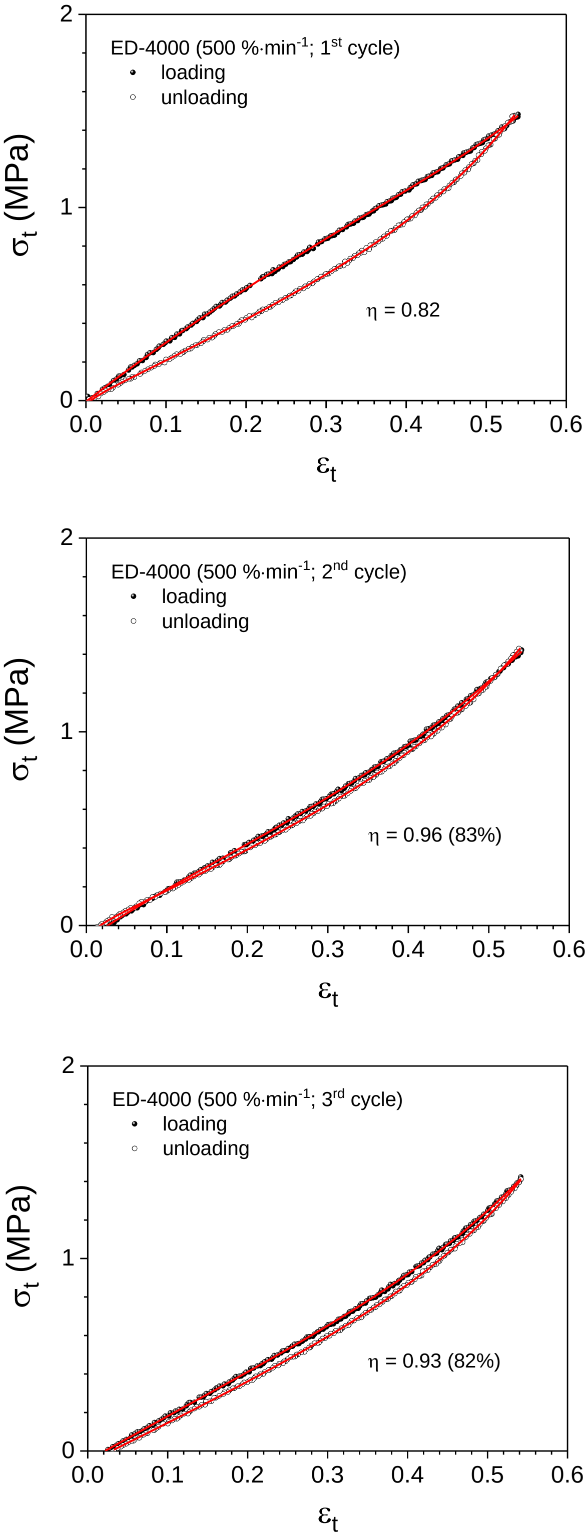


Figure SI-11. Uniaxial tensile test experiments for the PU elastomers ED-4000 during three loading-unloading cycles at the strain rate of 500 % min^{-1} with the corresponding fitting curves (red)

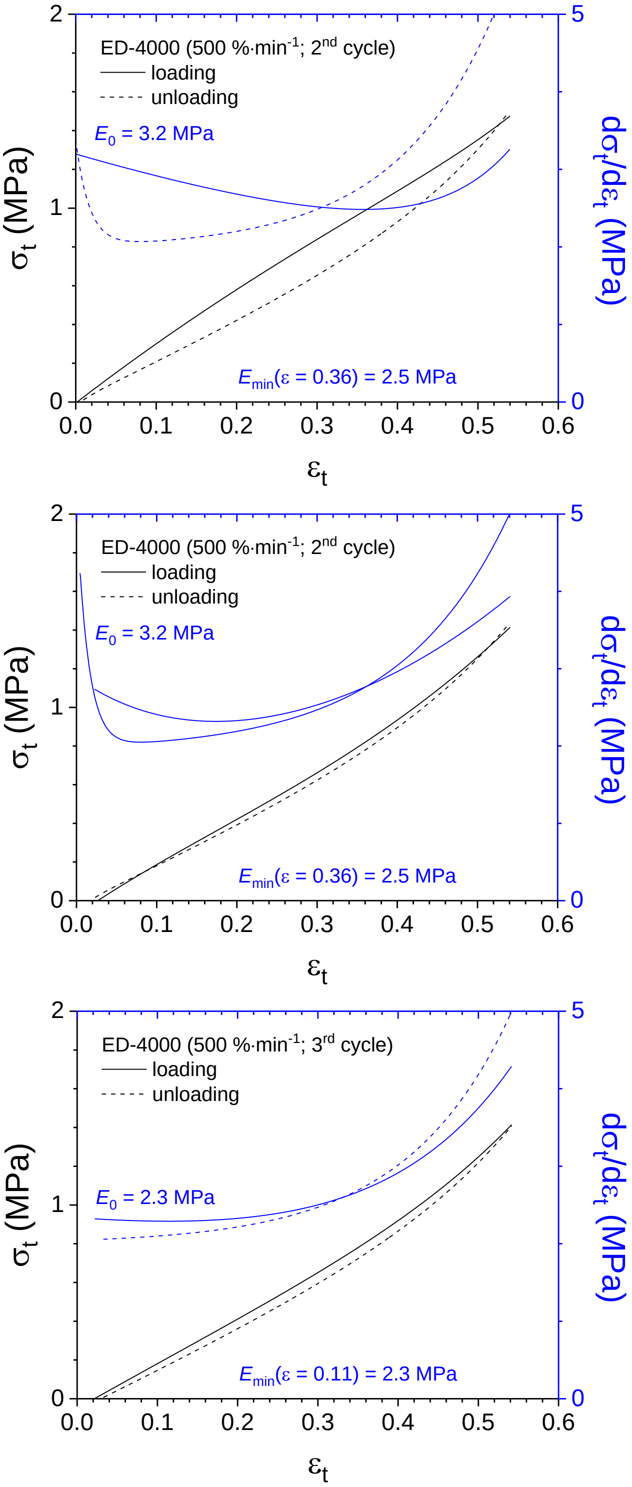


Figure SI-12. Fitting curves (black) and the corresponding derivatives (blue) for the three loading-unloading cycles at the strain rate of 500 % min^{-1} for the PU elastomers ED-4000.

Table SI-3. Fitting parameters obtained from the uniaxial tensile test experiments for the three PU elastomers ED-400, ED-2000 and ED-4000 during the three loading-unloading cycles at the strain rate of 500 % min^{-1}.

Sample	**Cycle #**	**a**	**b**	**c**	**d**	**f**
ED-400	1st loading	-	3.0	1.4	1.7	23
	unloading	5.5	0.0046	12.2	-	-
	2nd loading	-	0.18	5.9	1.7	5.6
	unloading	5.5	0.0056	12	-	-
	3rd loading	-	0.10	6.8	2.3	3.4
	unloading	5.6	0.0057	11	-	-
ED-2000	1st loading	-	1.8	1.6	0.50	6.1
	unloading	3.6	0.015	7.6	0.35	87
	2nd loading	1.8	0.15	0.41	0.68	1.3
	unloading	0.046	0.0014	0.087	0.0059	3.9
	3rd loading	-	0.096	5.0	3.8	0.93
	unloading	3.4	0.024	6.8	0.049	5.8
ED-4000	1st loading	-	0.00056	10	3.3	0.95
	unloading	2.0	0.0057	8.1	0.023	60
	2nd loading	-	0.46	2.3	0.33	5.5
	unloading	1.9	0.0084	7.3	0.045	70
	3rd loading	-	0.020	5.8	3.5	0.64
	unloading	-	0.0065	7.6	26	0.0077

Table SI-4. Initial elastic modulus (E_0), minimum elastic modulus (E_{min}), hysteresis (η) and energy recovery (ΔU) obtained from the uniaxial tensile test experiments for the three PU elastomers ED-400, ED-2000 and ED-4000 during the three loading-unloading cycles at the strain rate of 500 % min^{-1}.

Sample	**Cycle #**	**E_0 (MPa)**	**E_{min} (MPa)**	**η**	**ΔU**
ED-400	1st	43.9	6.1	0.47	
	2nd	8.9	6.7	0.74	57%
	3rd	7.7	6.6	0.73	54%
ED-2000	1st	5.9	4.8	0.75	
	2nd	4.6	4.1	0.89	78%
	3rd	4.0	4.0	0.87	77%
ED-4000	1st	3.2	2.5	0.82	
	2nd	2.7	2.3	0.96	83%
	3rd	2.3	2.3	0.93	82%

Appendix B

Procedure for the determination of material's parameters in the constitutive model

Using an inverse method optimization procedure [1], the constitutive parameters were determined by fitting model simulations to the experimental true stress-true strain monotonic and cyclic tensile curves. For this purpose, the MCalibration® commercial software by Veryst Engineering was utilized. The calibration was carried out in a sequence scheme: the elastic modulus was determined from the initial region of the true stress-true strain curve in the $0 < \varepsilon < 0.05$ range (Figure 14). The initial shear flow resistance $\hat{\tau}$, the strain-rate dependence exponent m together with the evolution parameter for ξ were determined in the $0.05 < \varepsilon < 0.1$ range (Figure 14). Following the orientation hardening parameters, μ and λ_L, were determined from the true stress-true strain curves simultaneously in the $0.1 < \varepsilon < 0.5$ region (Figure 14). A final fine-tuning optimization run was carried out calibrating all the parameters simultaneously using the complete true stress-true strain curves set together. Poisson's ratio was determined using the DIC transverse strain data in the initial elastic region.

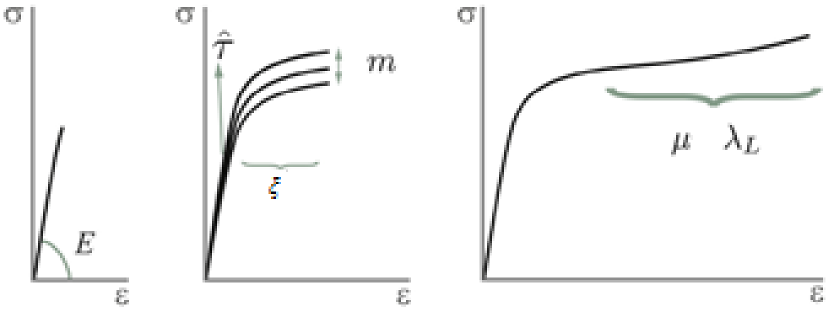


Figure SI-13. Schematic representing parameter calibration procedure.

REFERENCE

[1] Polanco-Loria M., Daiyan H., Grytten F.: Material parameters identification: An inverse modeling methodology applicable for thermoplastic materials. Polymer Engineering and Science, **52**, 438-448 (2012)

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