

Supplementally Material

## **Self- and Cross-fusing of Furan-based Polyurea Gels Dynamically Cross-linked with Maleimides**

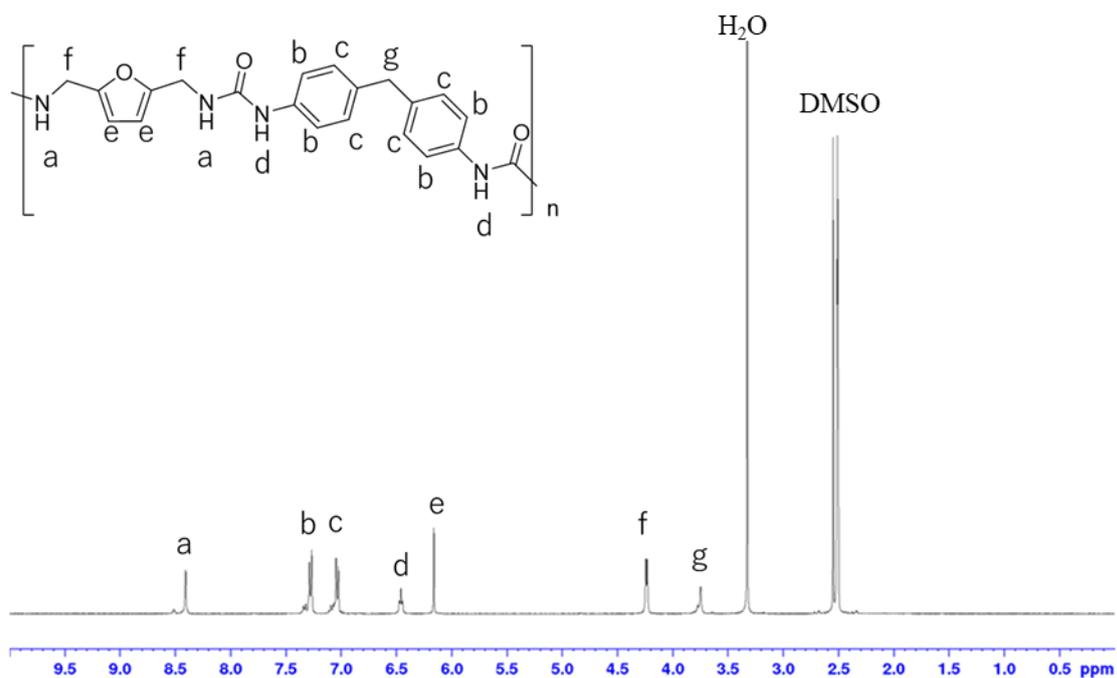
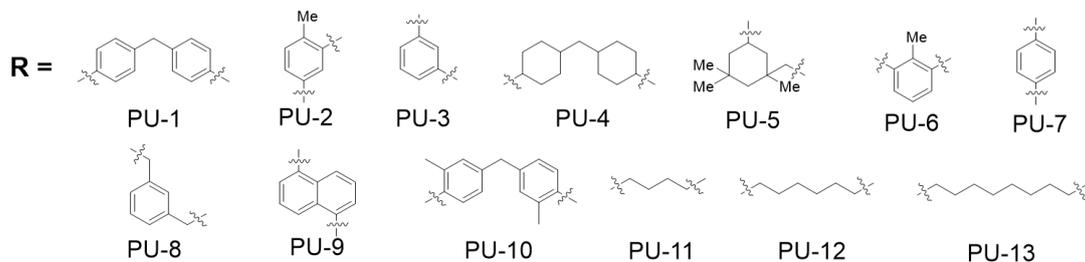
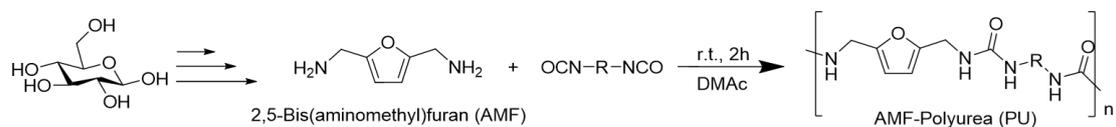
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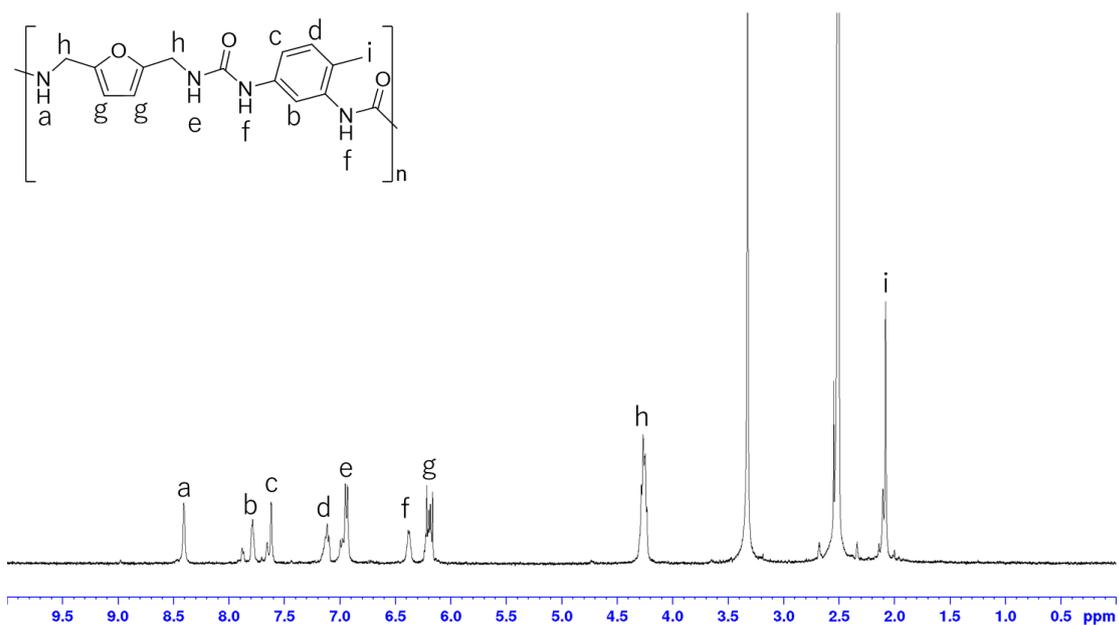
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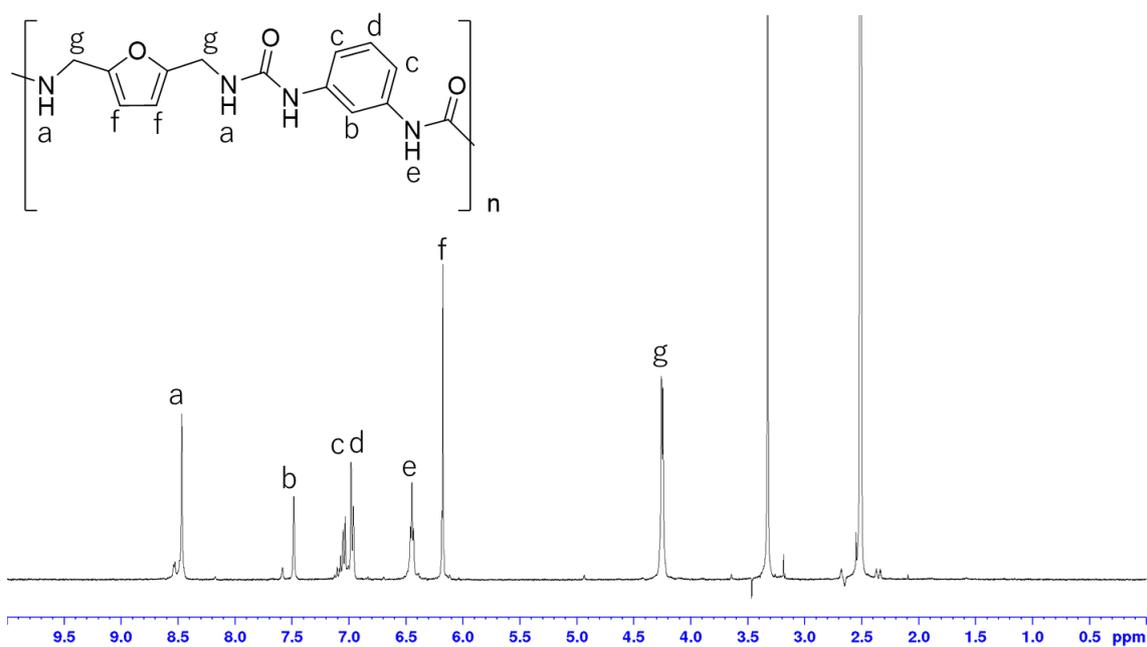
**Scheme S1.** Syntheses of polyureas from 2,5-bis(aminomethyl)furan and various diisocyanates.



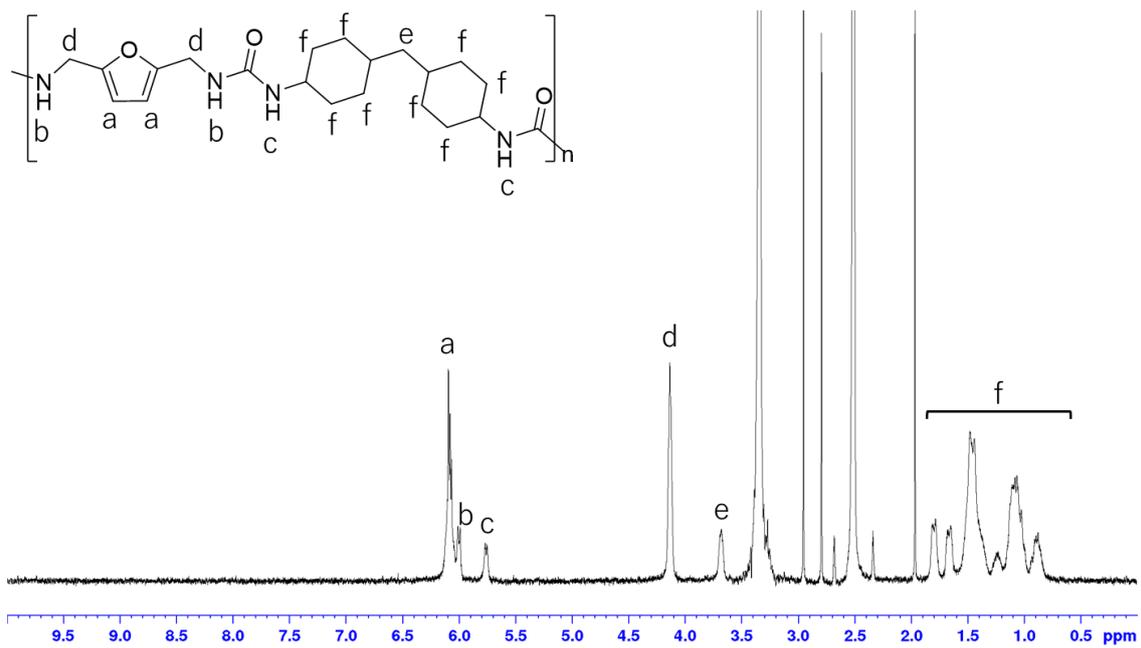
**Figure S1**  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ) spectrum of PU-1.



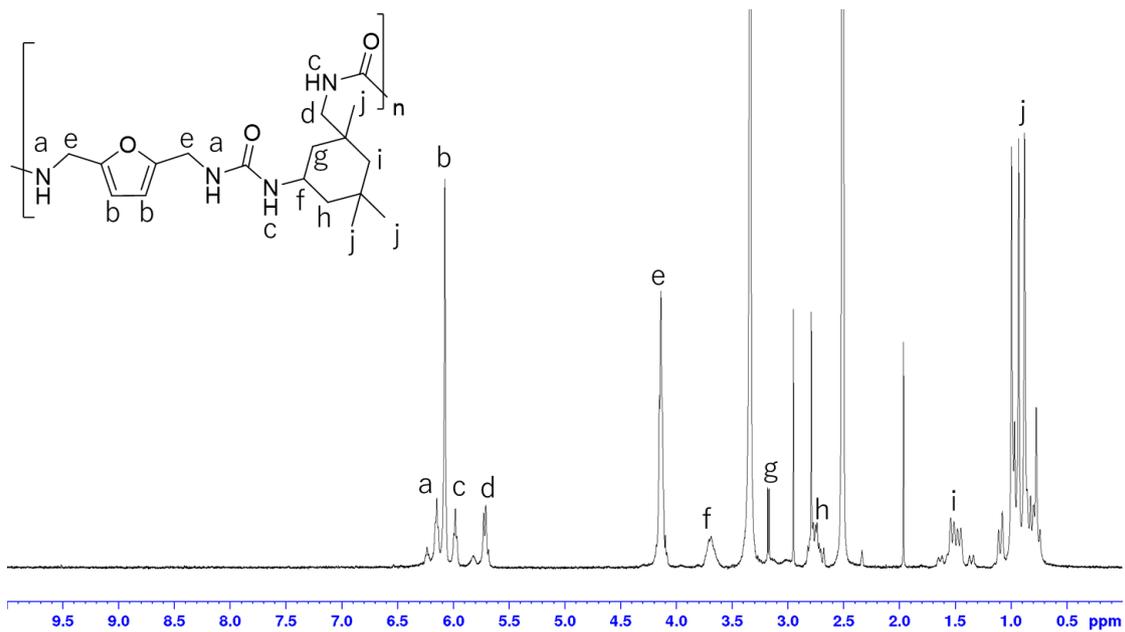
**Figure S2**  $^1\text{H NMR}$  (400 MHz,  $\text{DMSO-}d_6$ ) spectrum of PU-2.



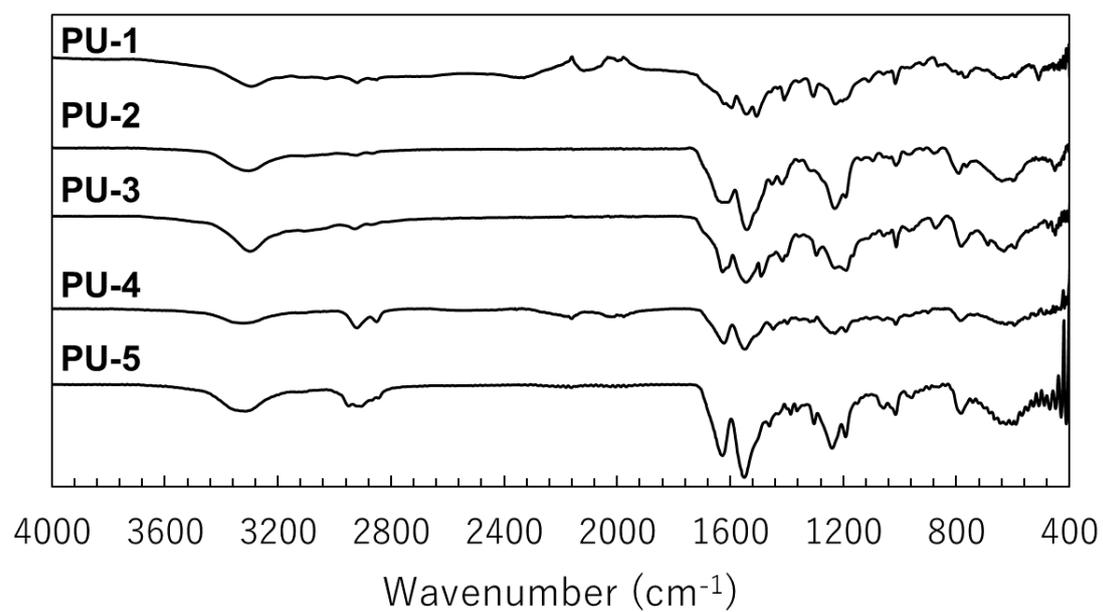
**Figure S3**  $^1\text{H NMR}$  (400 MHz,  $\text{DMSO-}d_6$ ) spectrum of PU-3.



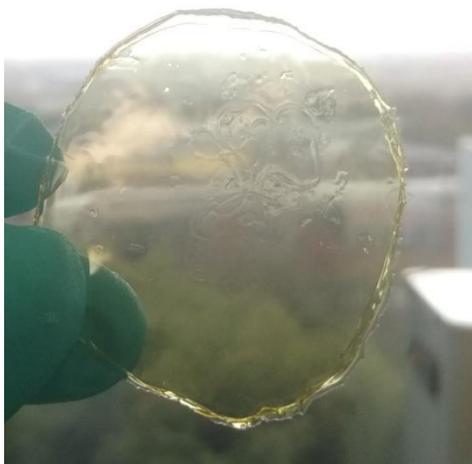
**Figure S4** <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) spectrum of PU-4.



**Figure S5** <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) spectrum of PU-5.



**Figure S6** IR spectra of PU-1 to PU-5 using AMF and various diisocyanates.



**Figure S7** Films of synthesized PU-1 using AMF.

**Table S1** Molecular weight, thermal, mechanical, and optical properties of the obtained PU-1 to 5

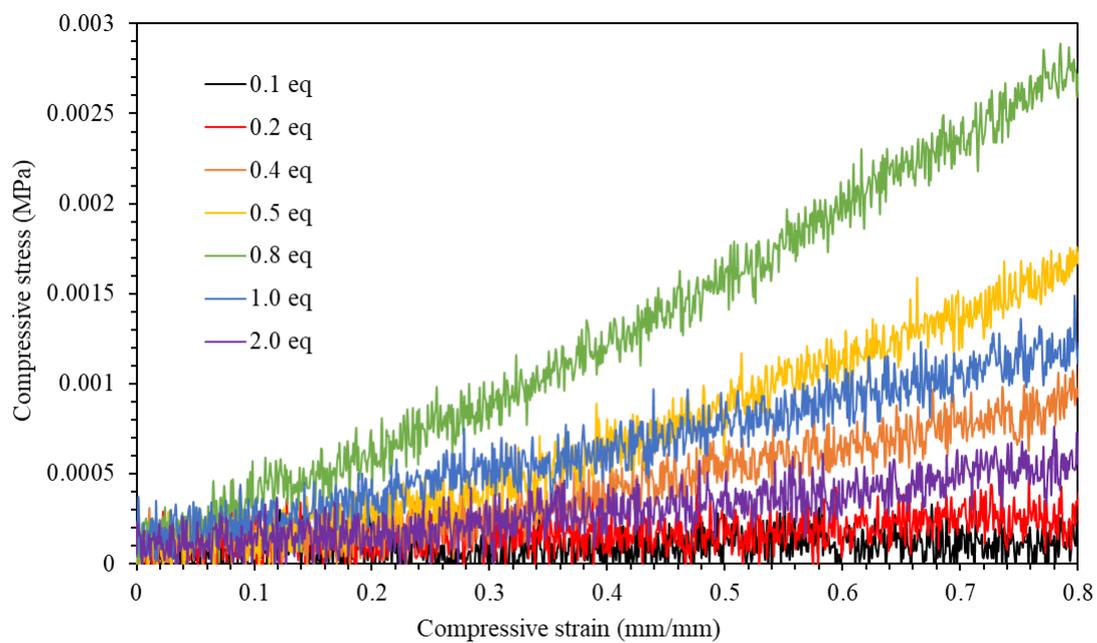
Polymer	$M_n^a$	$M_w^a$	$M_w/M_n^a$	$T_{d5} / ^\circ\text{C}^b$	$T_{d10} / ^\circ\text{C}^b$	$T_g / ^\circ\text{C}^c$	Strength, $\sigma / \text{MPa}$	Young's modulus, $E / \text{GPa}$	Elongation at break, $\varepsilon / \%$
PU-1	$7.4 \times 10^3$	$2.1 \times 10^4$	2.84	280	290	190	65	0.8	8.6
PU-2	$16 \times 10^3$	$94 \times 10^4$	6.01	280	290	<i>N.D.</i> <sup>f</sup>	25	0.5	6.8
PU-3	$8.8 \times 10^3$	$2.0 \times 10^4$	2.39	270	285	175	38	0.7	8.4
PU-4	$8.0 \times 10^3$	$2.0 \times 10^4$	2.65	310	320	210	16	0.3	8.9
PU-5	$3.0 \times 10^3$	$1.0 \times 10^4$	3.46	300	310	205	5	0.2	2.7

a) Determined by SEC measurement using DMF LiBr solution b) 5 % and 10 % weight loss temperatures.  $T_{d5}$  and  $T_{d10}$  were obtained from TGA curve scanned at a heating rate of 10 °C / min under  $\text{N}_2$  atmosphere. c)  $T_g$  was obtained from DSC curve scanned at a heating rate of 10 °C / min under  $\text{N}_2$  atmosphere. d) Not detected

**Table S2** Weight loss temperature and glass-transition temperature of the synthesized PUs using AMF and various diisocyanates.

	$T_{d1} (^\circ\text{C})^{(a)}$	$T_{d5} (^\circ\text{C})^{(a)}$	$T_{d10} (^\circ\text{C})^{(a)}$	$T_g (^\circ\text{C})^{(b)}$
PU-1	250	280	290	190
PU-2	240	280	290	<i>N.D.</i> <sup>(c)</sup>
PU-3	245	270	285	175
PU-4	290	310	320	210
PU-5	280	300	310	205
PU-6	245	270	280	<i>N.D.</i> <sup>(c)</sup>
PU-7	250	280	290	<i>N.D.</i> <sup>(c)</sup>
PU-8	270	295	300	150
PU-9	250	280	295	<i>N.D.</i> <sup>(c)</sup>
PU-10	255	290	300	<i>N.D.</i> <sup>(c)</sup>
PU-11	270	290	305	115
PU-12	280	300	310	<i>N.D.</i> <sup>(c)</sup>
PU-13	275	300	310	<i>N.D.</i> <sup>(c)</sup>

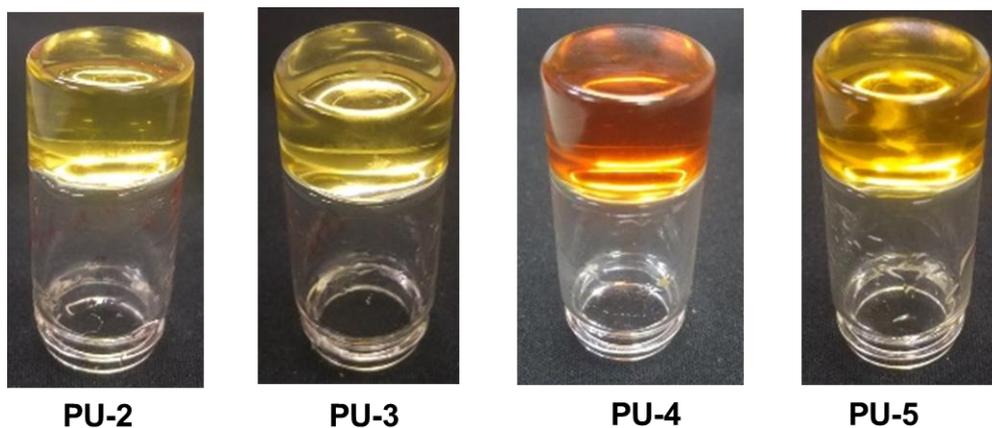
(a) 5 % and 10 % weight loss temperatures.  $T_{d1}$ ,  $T_{d5}$  and  $T_{d10}$  were obtained from TGA curve scanned at a heating rate of 10 °C / min under  $\text{N}_2$  atmosphere. (b)  $T_g$  was obtained from DSC curve scanned at a heating rate of 10 °C / min under  $\text{N}_2$  atmosphere. (c) Not detected



**Figure S8** Stress-strain curves of PU-1 gels with different of amount of BMI addition.

**Table S3** Mechanical properties of obtained PU-1 gels

Amount of BMI(mol%)	Elastic moduli $E_0$ (kPa)
0.1	$0.12 \pm 0.01$
0.2	$0.18 \pm 0.05$
0.4	$0.73 \pm 0.04$
0.5	$0.72 \pm 0.07$
0.8	$0.78 \pm 0.2$
1.0	$0.51 \pm 0.03$
2.0	$0.18 \pm 0.04$



**Figure S9** Photographs of gelation of various PUs

**Table S4** Mechanical properties of obtained polyurea gels and recovery of mechanical properties by DA reaction.

Amount of BMI(mol%)	Compressive strength, $\sigma$ (kPa)	Compressive strength after self-healing, $\sigma$ (kPa)	Compression strain, $\varepsilon$ (mm/mm)	Compression strain after self-healing, $\varepsilon$ (mm/mm)
10	17.2	23.2	0.9	0.8
50	39.4	60.5	0.6	0.6
80	23.9	15.5	0.5	0.4
100	33.6	38.4	0.6	0.6