

Supporting Information

Hydrogen Bond Reinforced, Transparent Polycaprolactone- based Degradable Polyurethane

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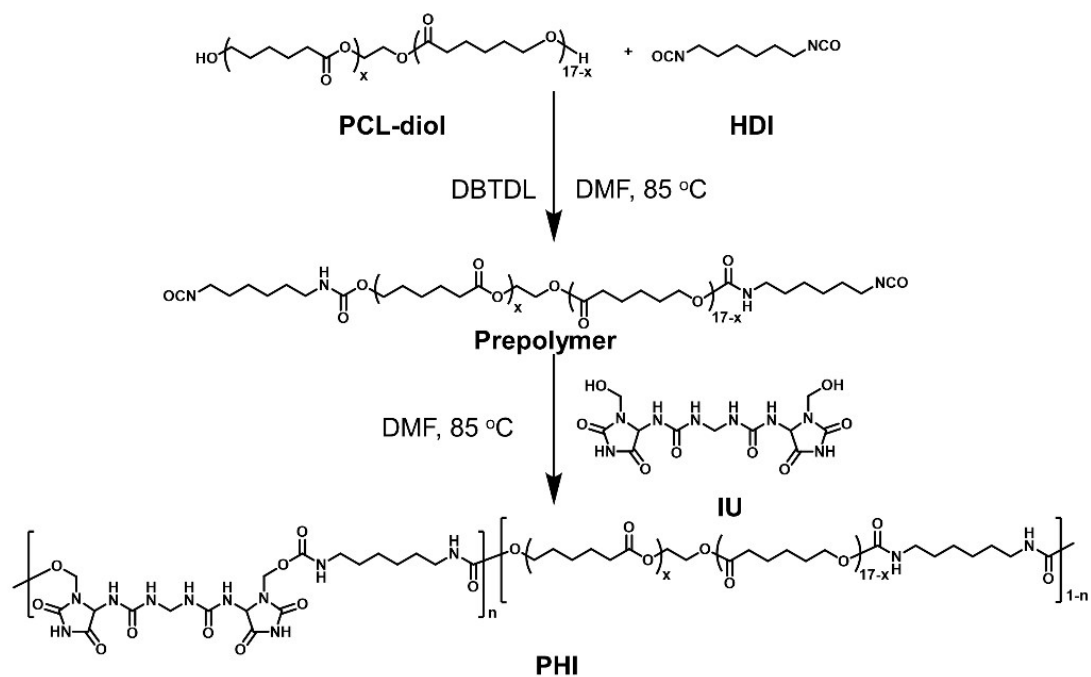


Figure S1. Synthetic routes of polymeric elastomers with hierarchical hydrogen bonding.

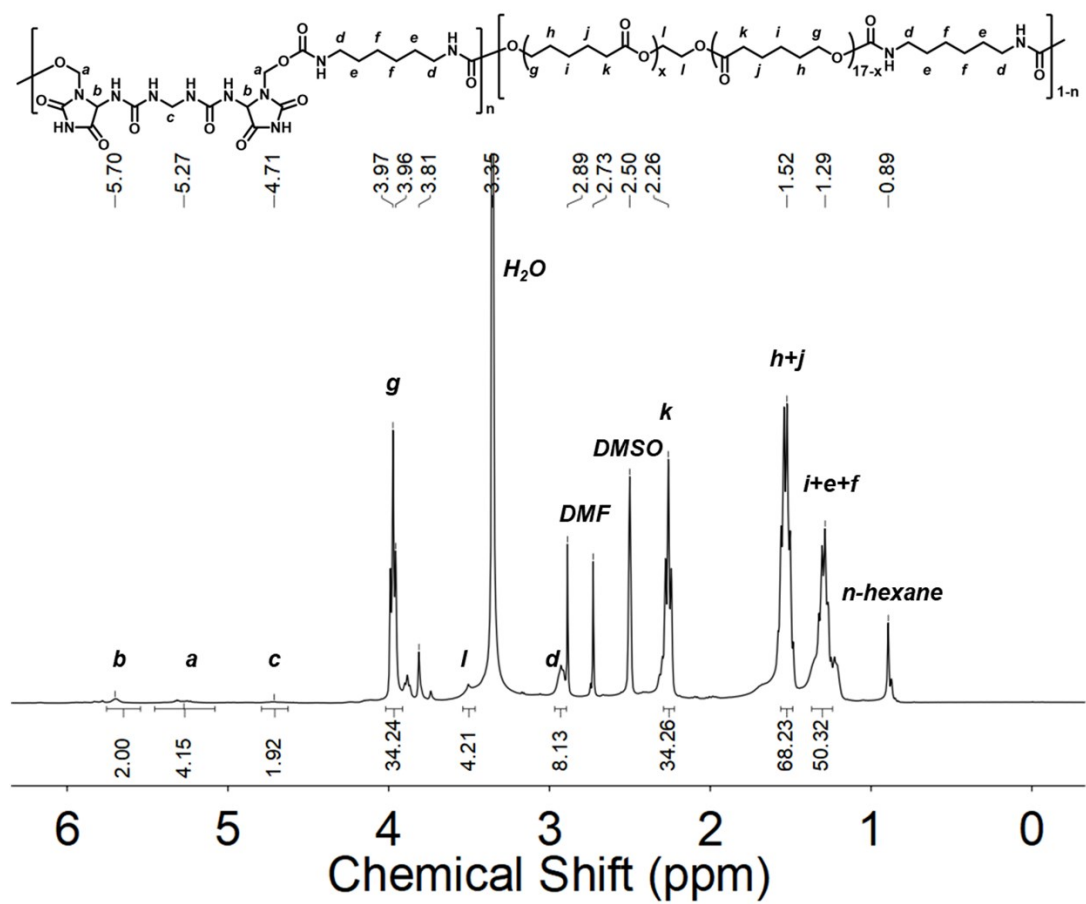


Figure S2. ^1H NMR spectra of sample PHI25.

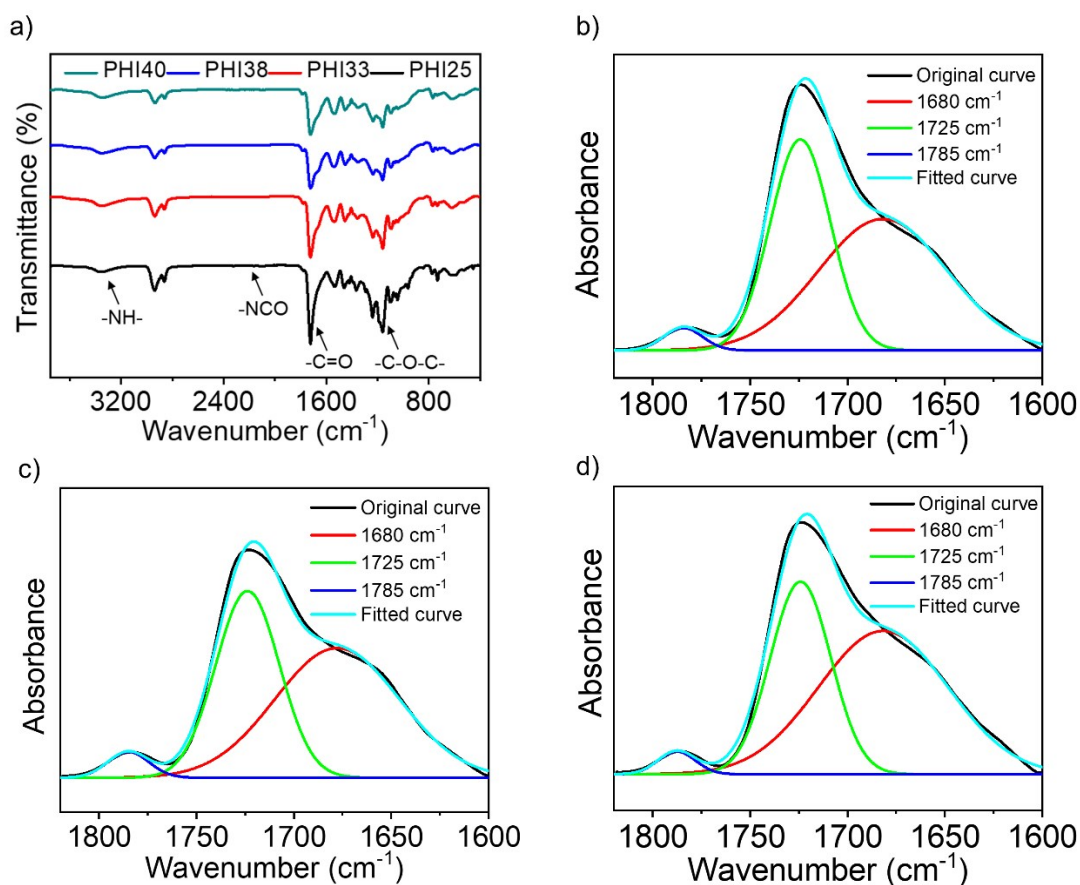


Figure S3. FTIR of PHI samples. a) The transmittance of all PHI films. b) The FTIR spectra of PHI33 films fitted by Gauss curves for 1850~1600 cm^{-1} . c) The FTIR spectra of PHI38 films fitted by Gauss curves for 1850~1600 cm^{-1} . d) The FTIR spectra of PHI40 films fitted by Gauss curves for 1850~1600 cm^{-1} .

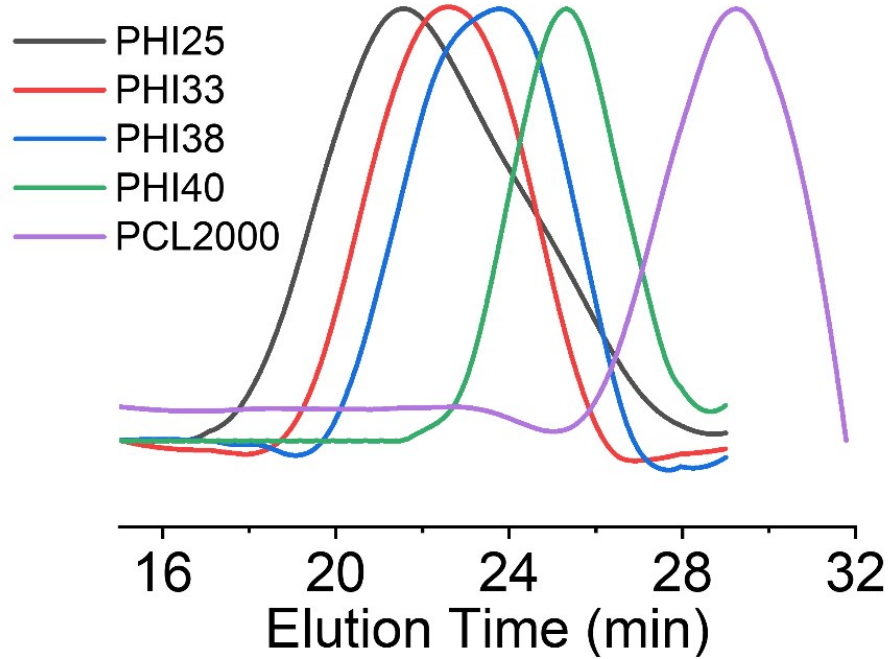


Figure S4. GPC curves of all PHI samples.

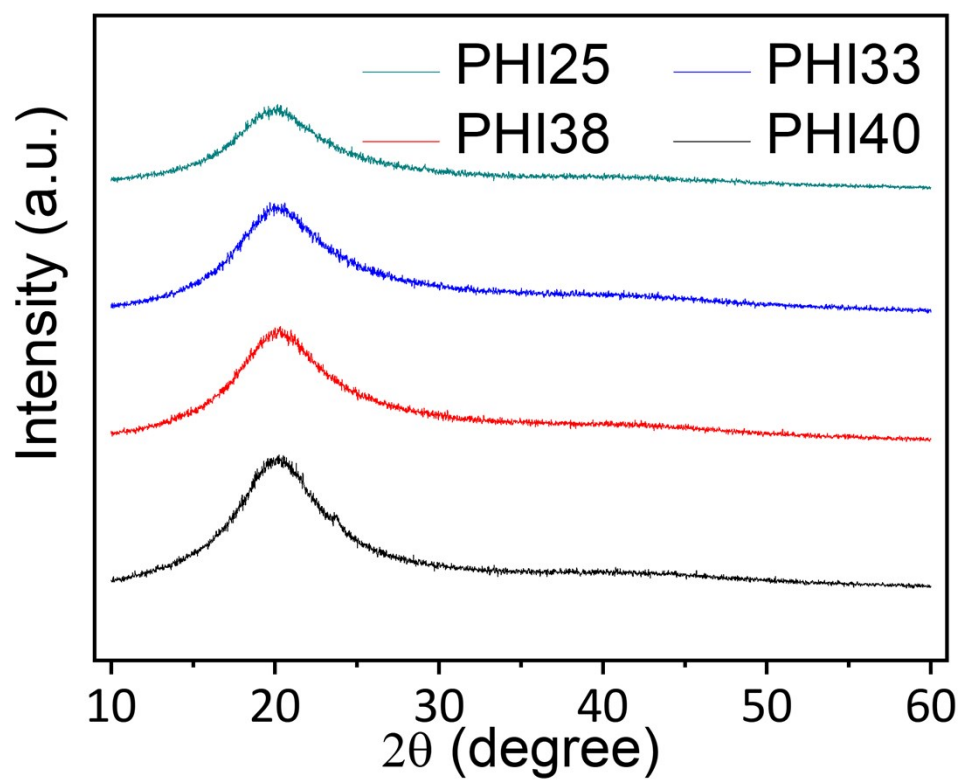


Figure S5. XRD curves for all PHI films.

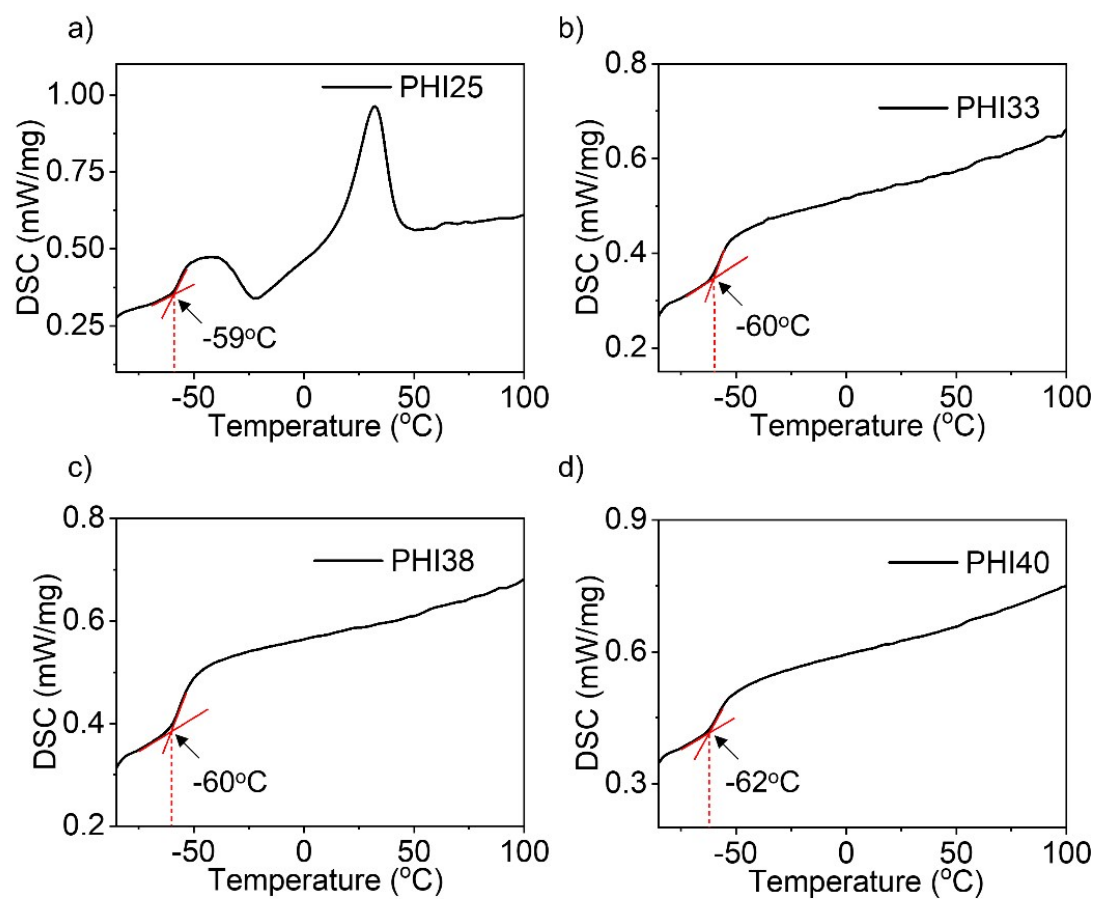


Figure S6. DSC of all PHI samples.

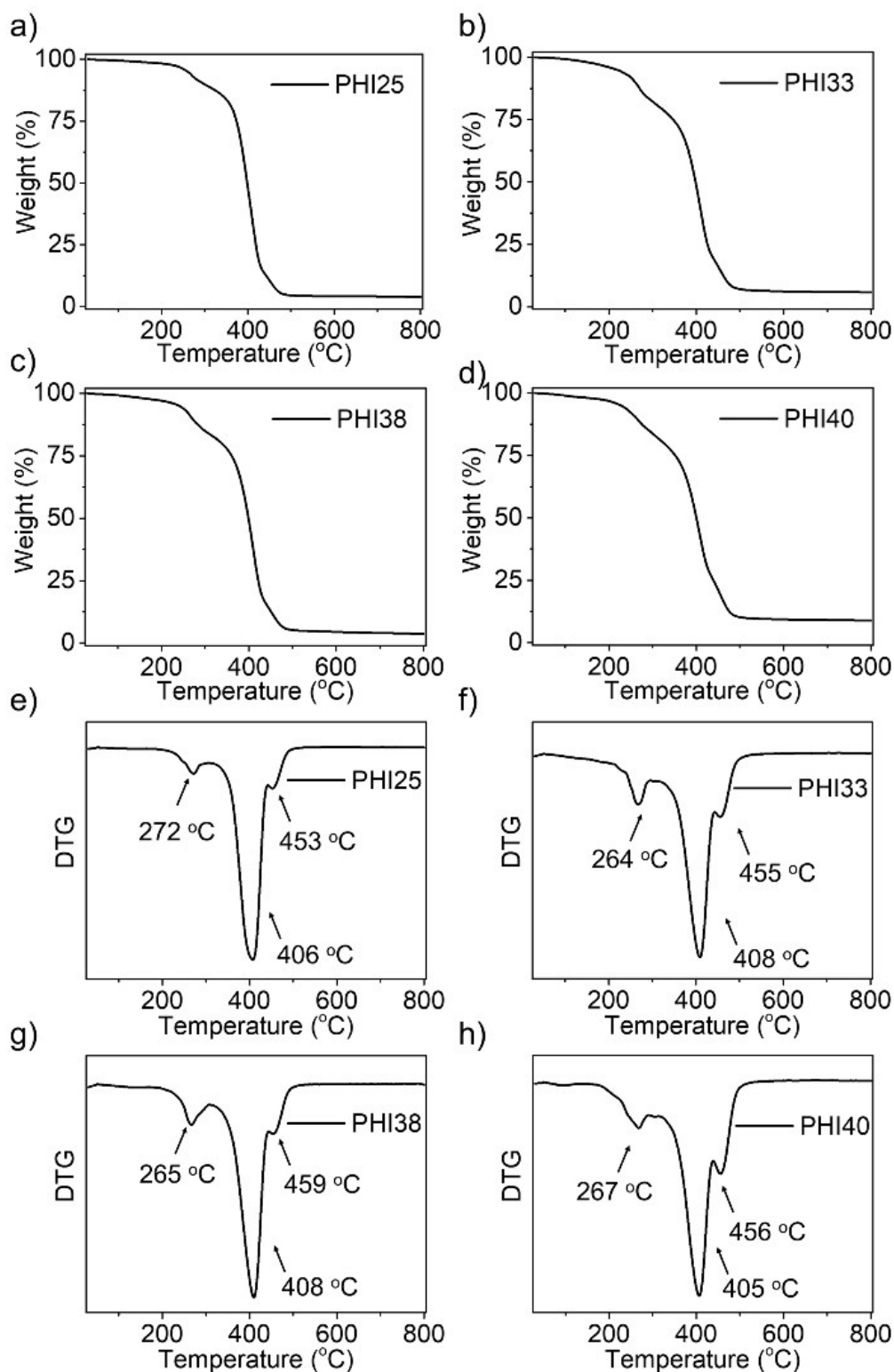


Figure S7. TGA of all PHI films. a ~ d) Thermogravimetric analysis of all PHI samples.

e ~ f) DTG curves of all PHI samples.

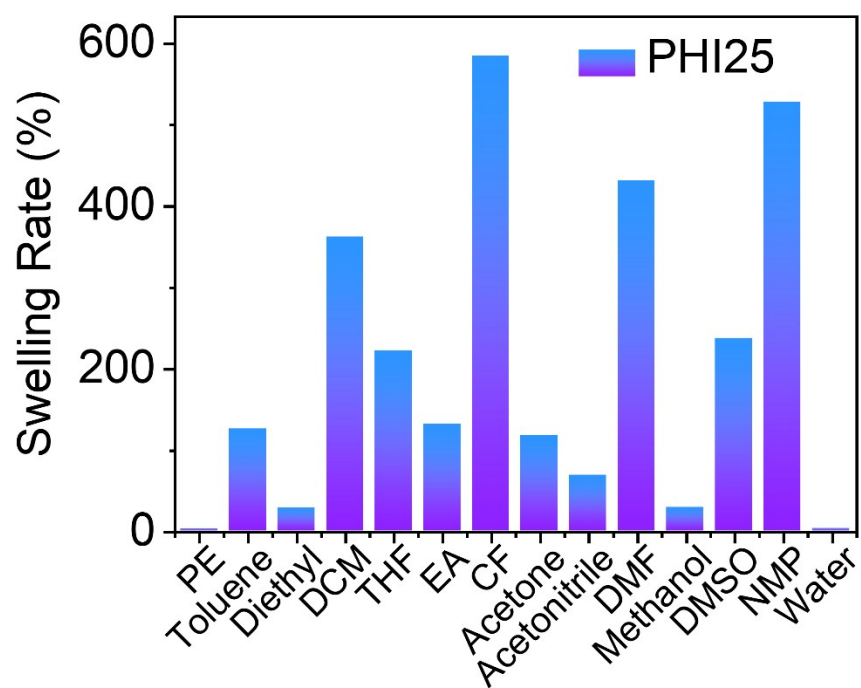


Figure S8. Swelling rate of PHI25 films in different polar solvents.

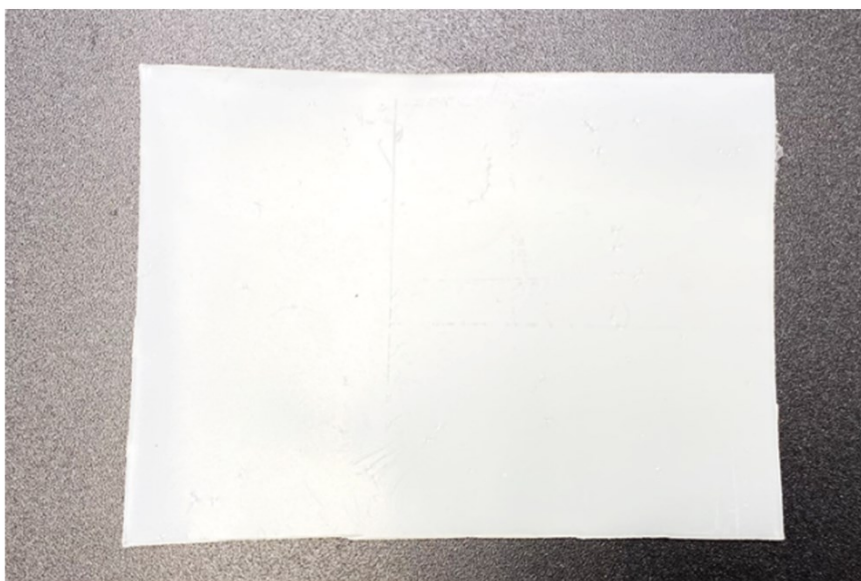


Figure S9. The photo of PHI0.

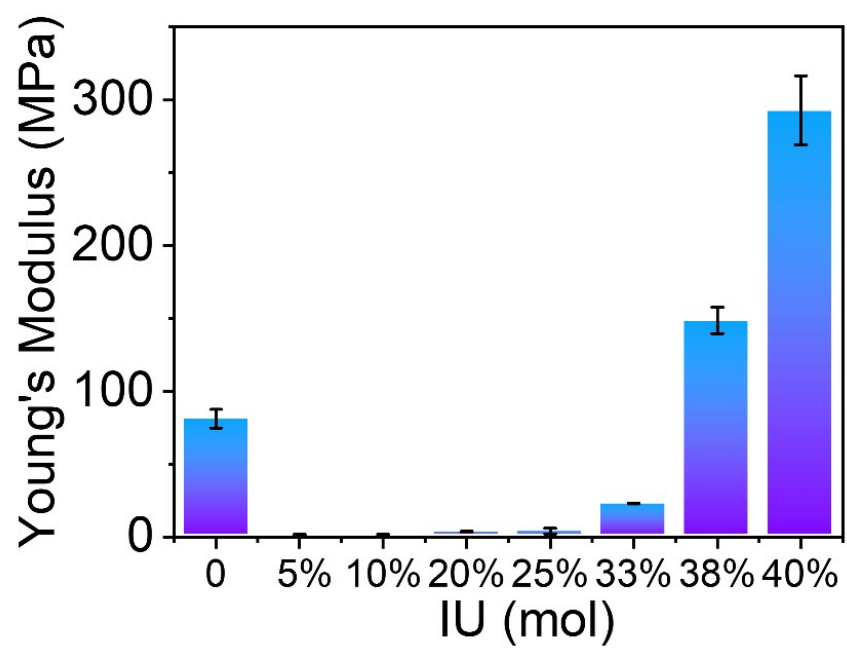


Figure S10. Young's modulus of PHI.

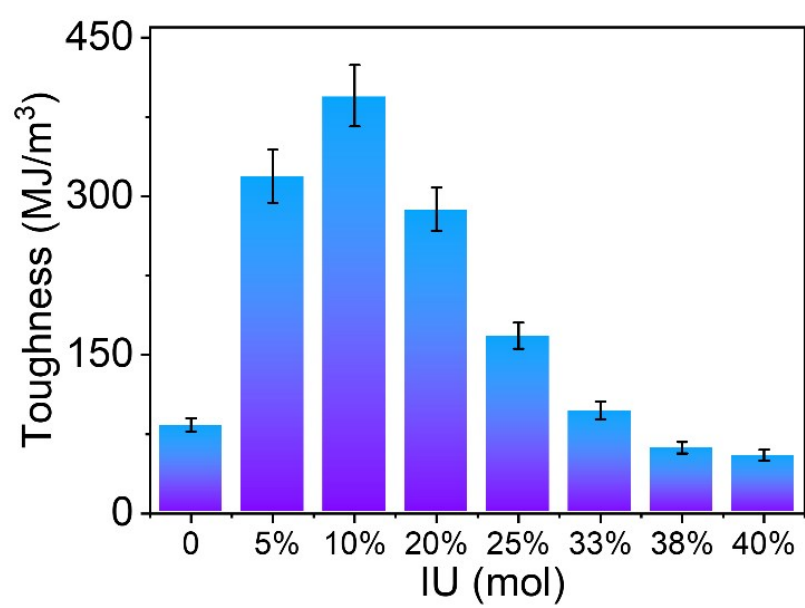


Figure S11. The toughness of PHI samples.

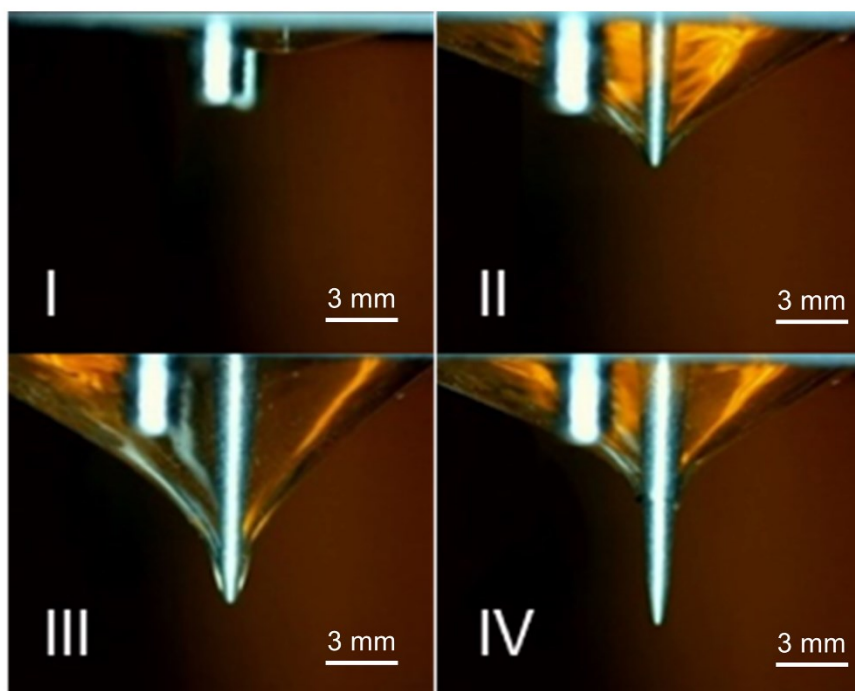


Figure S12. Images showing puncture tests of sample PHI25 with different displacements.

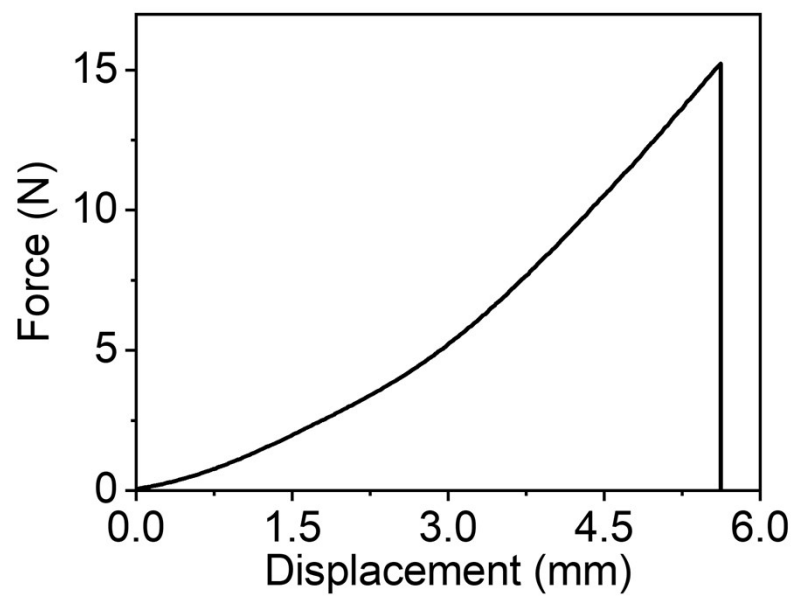


Figure S13. Typical puncture force-displacement curve of sample PHI33.

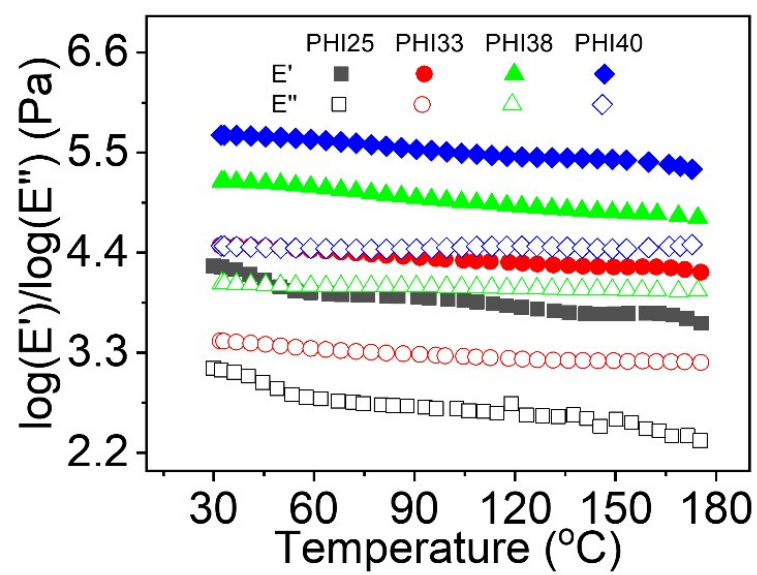


Figure S14. Temperature dependence of the storage modulus E' and the loss modulus E'' of PHI.

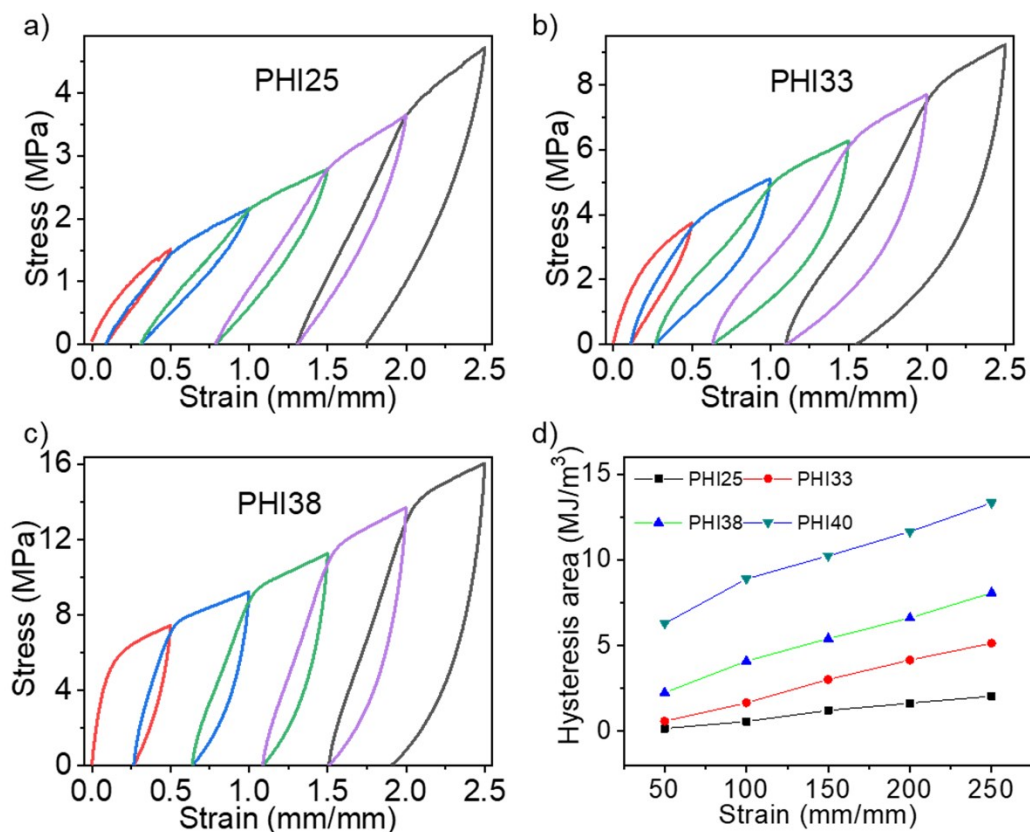


Figure S15. Step-cycle tensile deformation. a~c) Loading-unloading curves of samples PHI25~PHI38 under different strains. d) Dissipated energy during loading-unloading processes under different strains.

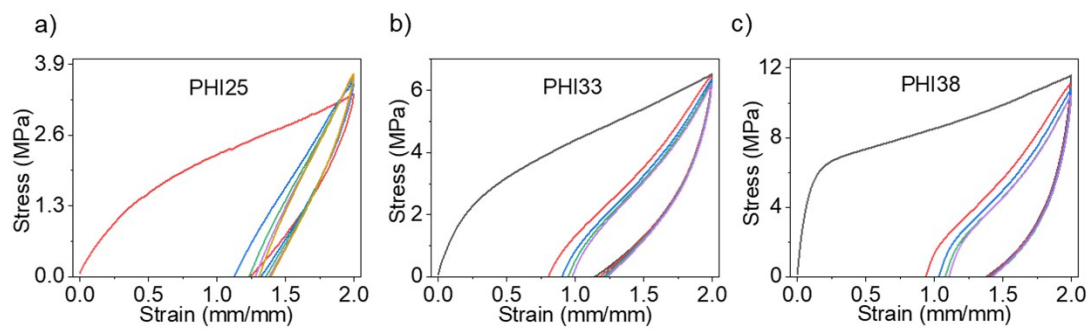


Figure S16. Five successive loading-unloading cycles with 200% strain of samples PHI25~PHI38.

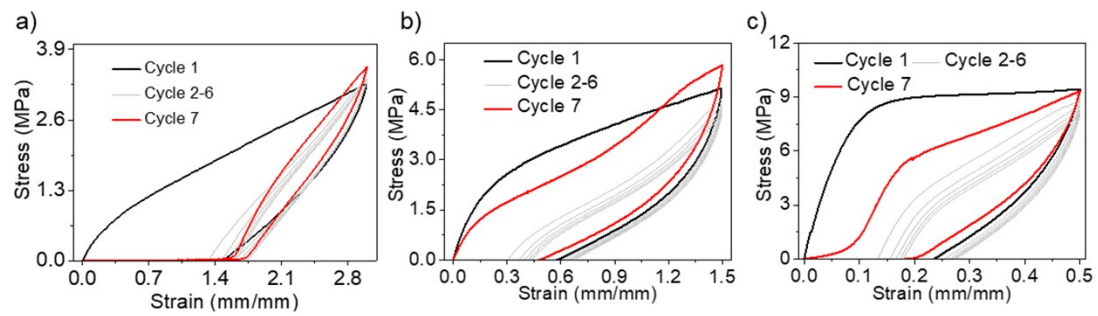


Figure S17. Cyclic tensile test curves in successive cycles of samples PHI25~PHI38.

The loading-unloading cycle 7 was delayed for 2 h after cycle 6.

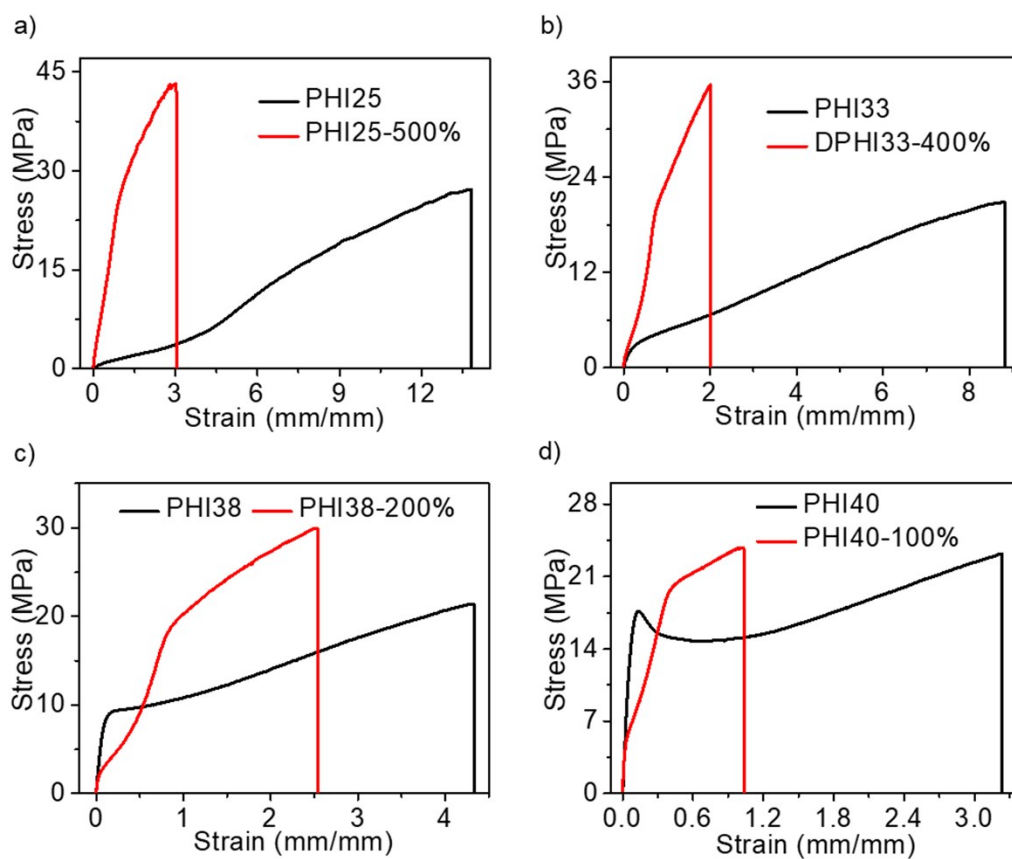


Figure S18. Monotonic stress-strain curves of stretch-reinforced elastomer. The black line is the curve of pristine sample and the red line is the sample deformed to a different strain.

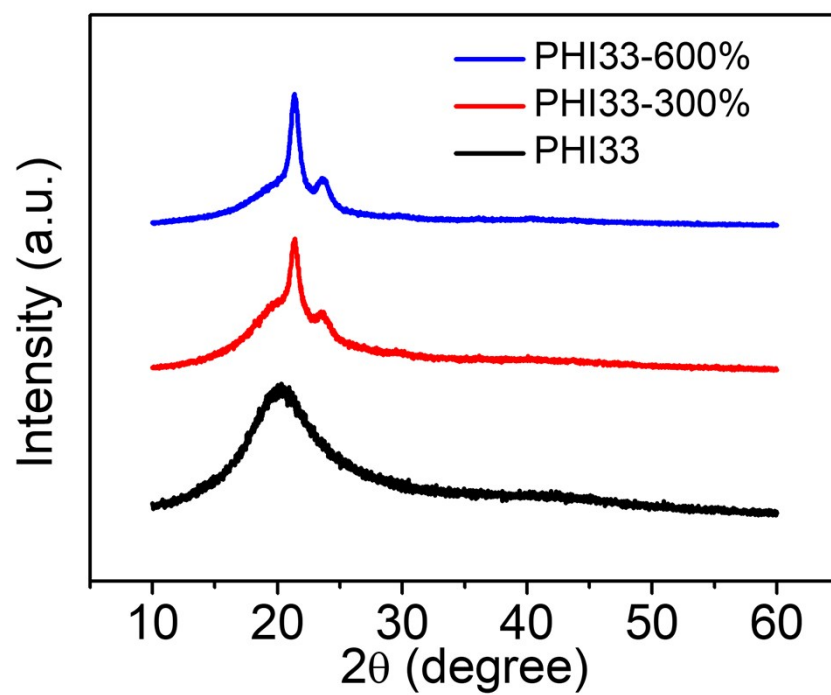


Figure S19. Wide-angle X-ray scattering of sample PHI33 at different deformation.

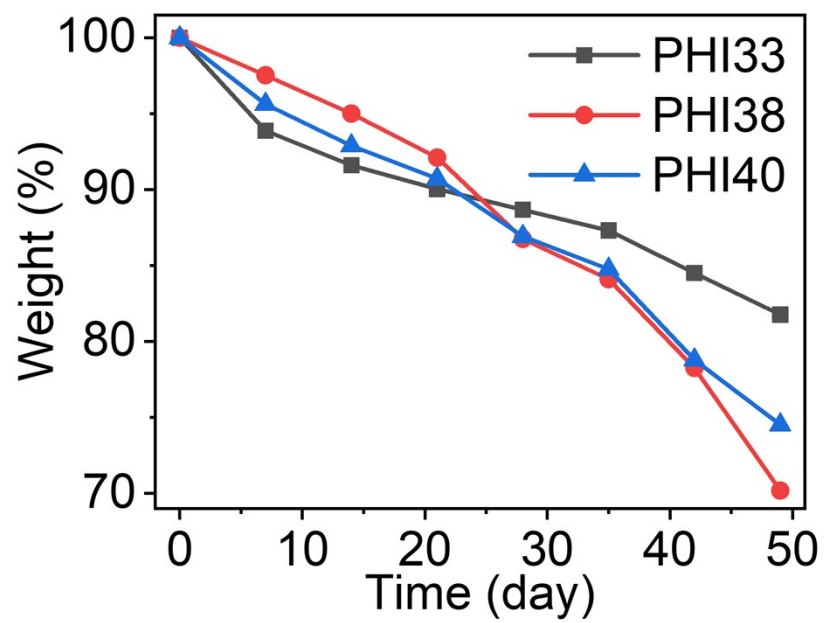


Figure S20. The degradation curves of all samples.

Table S1. Synthesis parameters of PHI.

Sample	PCL2000 (mol)	HDI (mol)	IU (mol)	IU (mol%)	Mn (KDa)	Mw (KDa)	PDI
PHI25	1	2	1	25	31.6	41.1	1.3
PHI33	1	3	2	33	24.7	29.6	1.2
PHI38	1	4	3	38	19.3	23.2	1.2
PHI40	1	5	4	40	11.8	13.0	1.1
PCL2000	/	/	/	/	2.0	2.2	1.1

Table S2. Mechanical properties of PHI samples.

IU/mol%	Modulus (MPa)	Breaking Strength (MPa)	Breaking Strain (mm/mm)	Toughness (MJ/m ³)
25	4.1 ± 1.9	24.9 ± 2.3	13.8 ± 0.2	168.2 ± 22.1
33	23.0 ± 0.2	19.9 ± 0.9	8.4 ± 0.6	97.5 ± 11.2
38	148.7 ± 9.0	21.1 ± 0.8	4.1 ± 0.2	62.2 ± 4.2
40	293.0 ± 23.6	19.8 ± 4.3	3.3 ± 0.4	55.2 ± 6.6

Table S3. Mechanical properties of sample PHI25 with different pre-stretched deformation.

Deformation mm/mm	Original Length (mm)	The Length of Residual Deformation (mm)	Modulus (MPa)	Breaking Strength (MPa)	Breaking Strain (mm/mm)
0	16.00	16.00	4.1 ± 1.9	24.9 ± 2.3	13.8 ± 0.2
200%	16.00	25.15 ± 1.99	4.1 ± 0.4	23.1 ± 1.1	7.4 ± 0.9
400%	16.00	58.69 ± 2.14	25.8 ± 3.0	35.9 ± 5.3	2.4 ± 0.4
600%	16.00	70.52 ± 8.73	49.9 ± 12.0	48.5 ± 5.7	2.0 ± 0.7

Table S4. Mechanical properties of PHI samples with different deformation rates.

Sample	Modulus (MPa)	Breaking Strength (MPa)	Breaking Strain (mm/mm)	Toughness (MJ/m ³)
PHI25	4.1 ± 1.9	24.9 ± 2.3	13.8 ± 0.2	168.2 ± 22.1
PHI25-500%	34.6 ± 0.3	43.9 ± 4.1	3.0 ± 0.3	88.7 ± 0.8
PHI33	23.0 ± 0.2	19.9 ± 0.9	8.4 ± 0.6	97.5 ± 11.2
PHI33-400%	47.5 ± 0.8	34.5 ± 1.2	2.1 ± 0.08	42.4 ± 0.01
PHI38	148.7 ± 9.0	21.1 ± 0.8	4.1 ± 0.2	62.2 ± 4.2
PHI38-200%	103.4 ± 6.0	26.8 ± 3.2	2.3 ± 0.2	50.0 ± 0.4
PHI40	293.0 ± 23.6	19.8 ± 4.3	3.3 ± 0.4	55.2 ± 6.6
PHI40-100%	253.9 ± 7.6	22.4 ± 1.4	1.1 ± 0.1	18.5 ± 0.2

Video S1. Puncture Test of PHI. (MP4)