Supporting Information

A Highly Stretchable, Transparent, Notch-insensitive Self-Healing Elastomers for Coating

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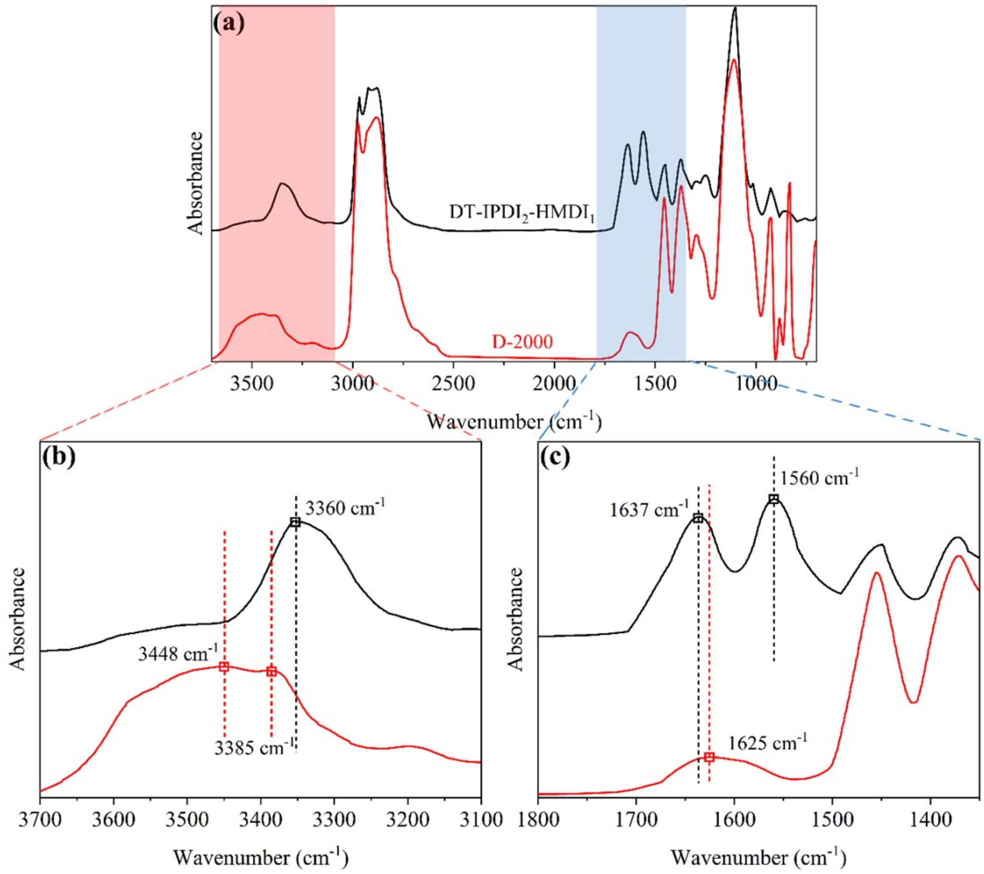

Fig. S1 The FT-IR spectrum of polyether amine D-2000 (red) and polyurea elastomer DT-IPDI$_2$-HMDI$_1$ (black) in the range of 600-3600 cm^{-1} (a); urea group (1350-1800 cm^{-1}) stretching zone of DT-IPDI$_2$-HMDI$_1$ and N-H Bending vibration (1350-1800 cm^{-1}) of D-2000 (b); N-H (3700-3100 cm^{-1}) stretching zone (c).

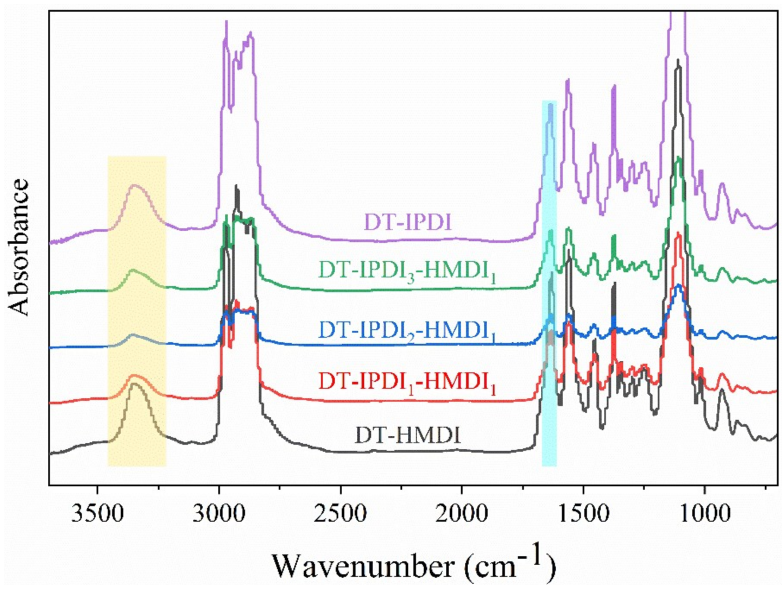

Fig. S2 Comparative FT-IR spectra of DT-IPDI$_x$-HMDI$_y$. The appearance of C=O at 1640 cm^{-1} (blue region) and N-H at 3360 cm^{-1} (yellow region) indicate the success synthesis of DT-IPDI$_x$-HMDI$_y$ elastomers.

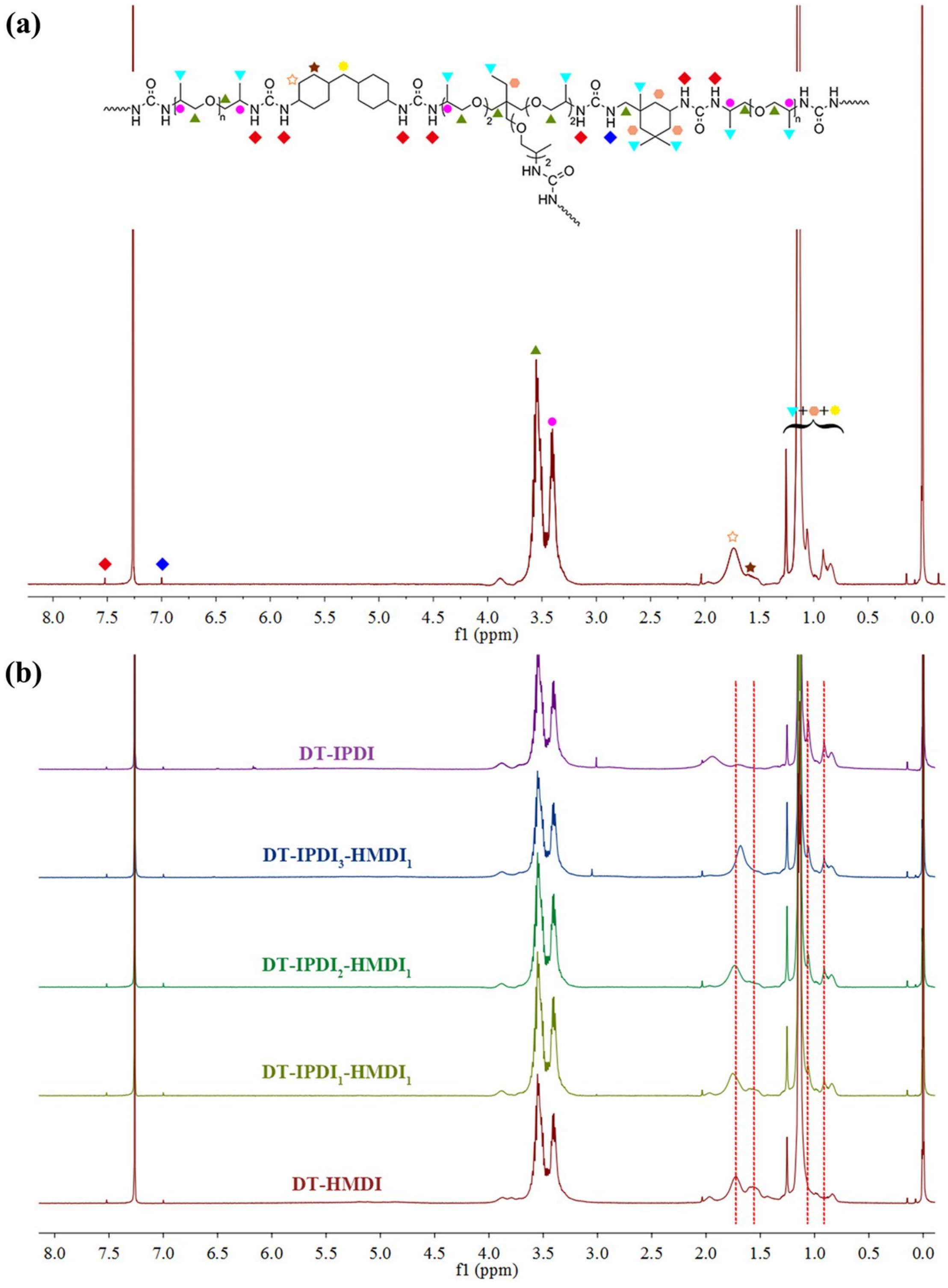

Fig. S3 (a) ^{1}H NMR spectra of the DT-IPDI$_2$-HMDI$_1$. (b) Comparative ^{1}H NMR spectra of polyurea elastomers at different IPDI-HMDI ratios. the color of the spectral curve is consistent with the labeling color of the formula.

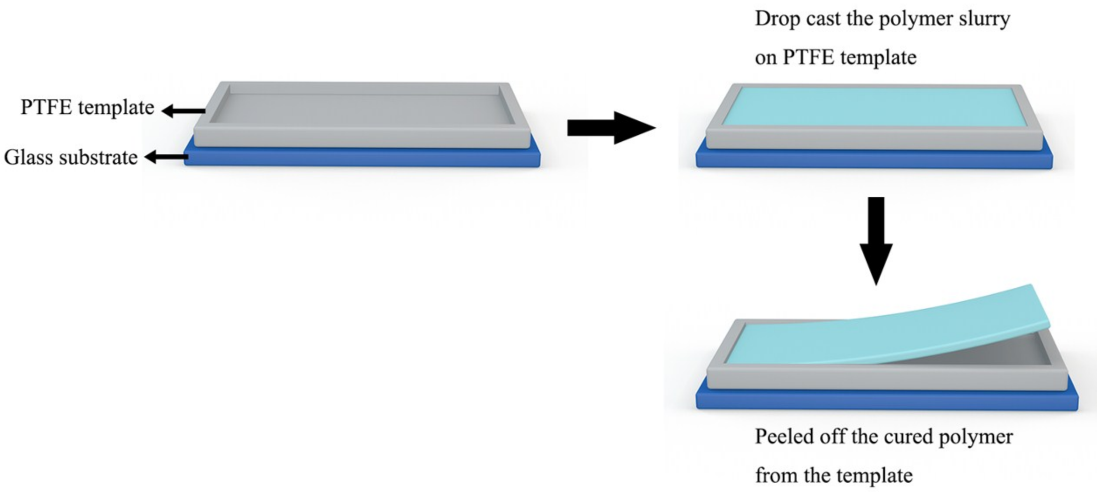

Fig. S4 Flow chart for the preparation of sample films used for testing.

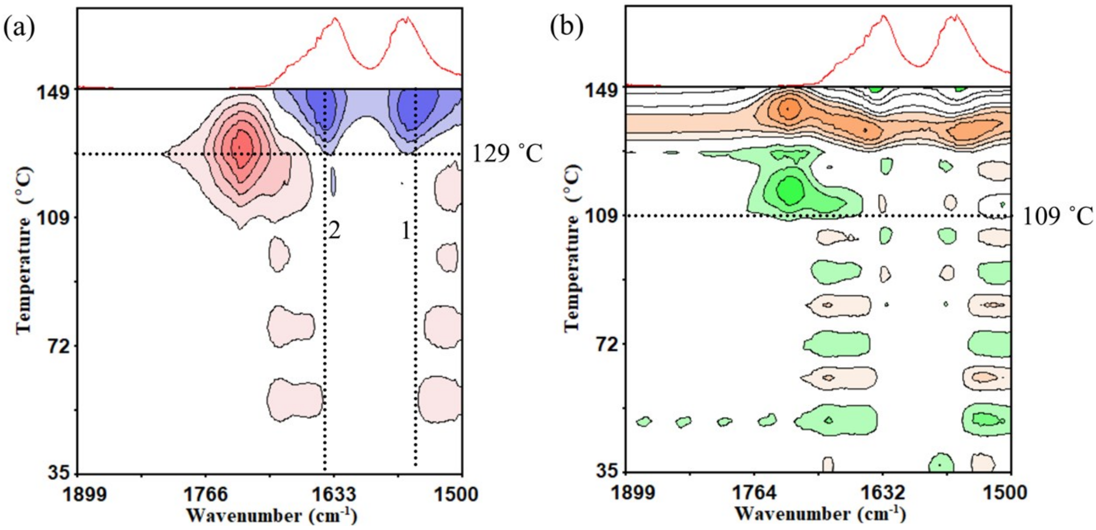

Fig. S5 (a) PCMW2D synchronous and (b) asynchronous FTIR spectra of DT-IPDI$_2$-HMDI$_1$ elastomer upon heating. The pink (orange) areas represent positive correlation intensity, while blue (green) areas represent negative correlation intensity.

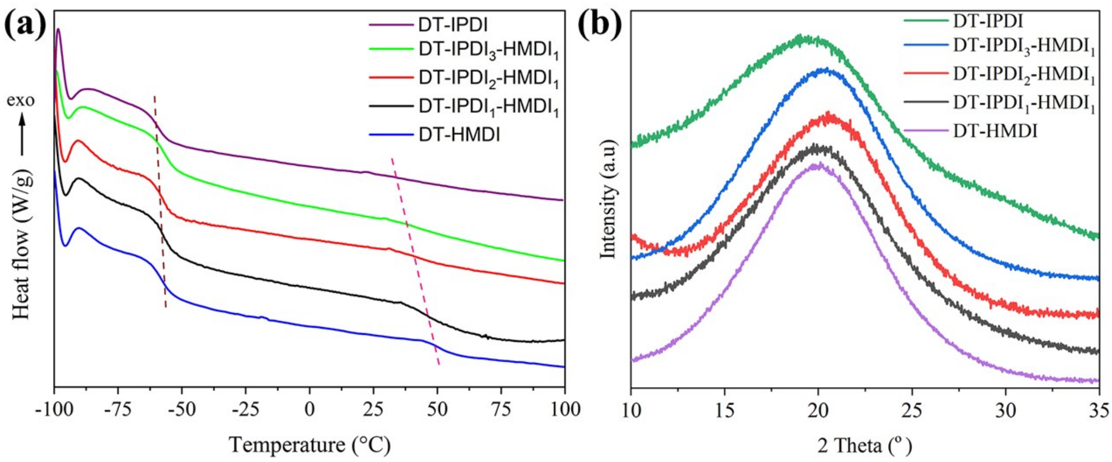

Fig. S6 (a) DSC heating curves of DT-IPDI$_x$-HMDI$_y$ was collected during the second heating process from −100 to 100 ºC at a fixed rate of 10 ºC/min. (b) WAXS curves of DT-IPDI$_x$-HMDI$_y$ samples.

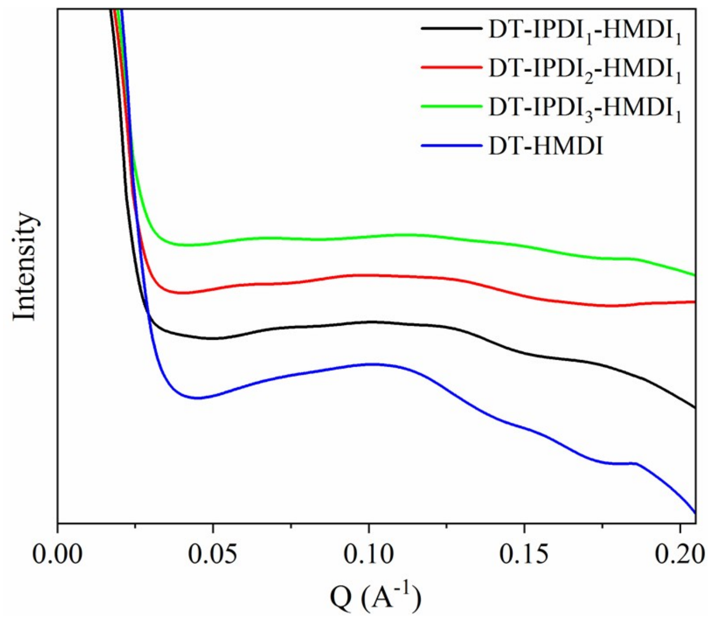

Fig. S7 SAXS profile plot for DT-IPDI$_x$-HMDI$_y$ that supports the presence of microphase separation.

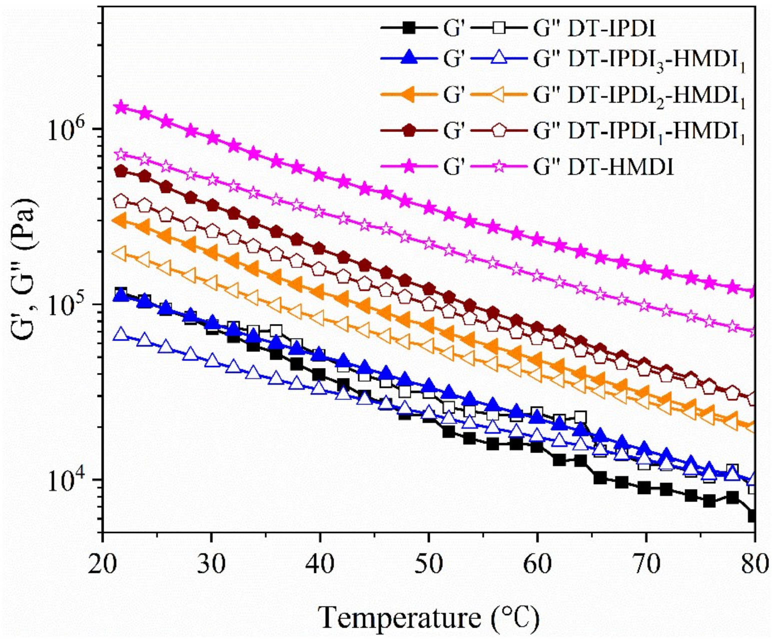

Fig. S8 Temperature-dependent rheological curve of DT-IPDIx-HMDIy at 1Hz.

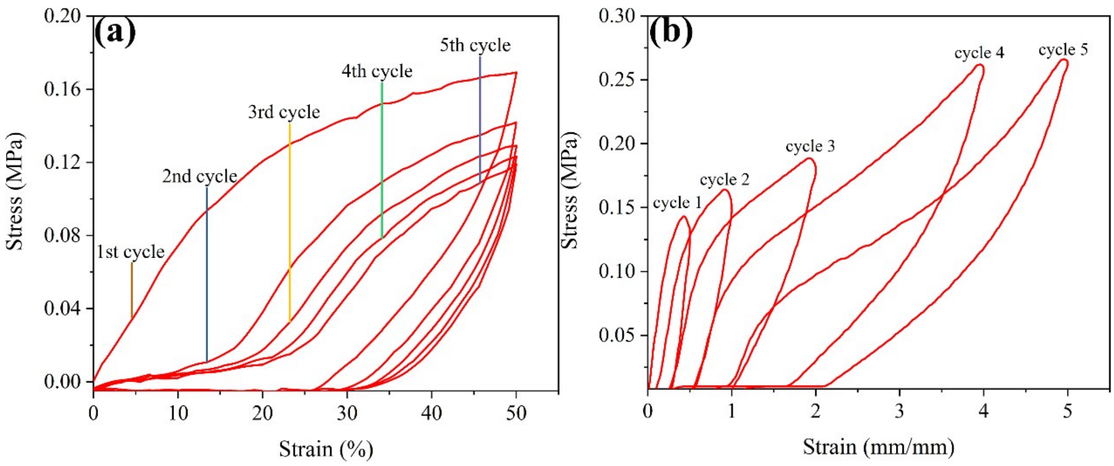

Fig. S9 (a) Recovery and cyclic loading of the dumbbell spline DT-IPDI$_2$-HMDI$_1$. The samples were loaded (50% strain) and immediately released 5 times (tensile rate: 100 mm/min). (b) Cyclic tensile test of the DT-IPDI$_2$-HMDI$_1$ at a settled stretch rate 100 mm/min, the sample was loaded to 50% strain firstly and then released to the original state (cycle 1). After that stretched to a higher strain (100%) immediately released the load which marked cycle 2. The cyclic test was repeated as the similar procedure gradually increased the strain with 200%, 400% and 500%.

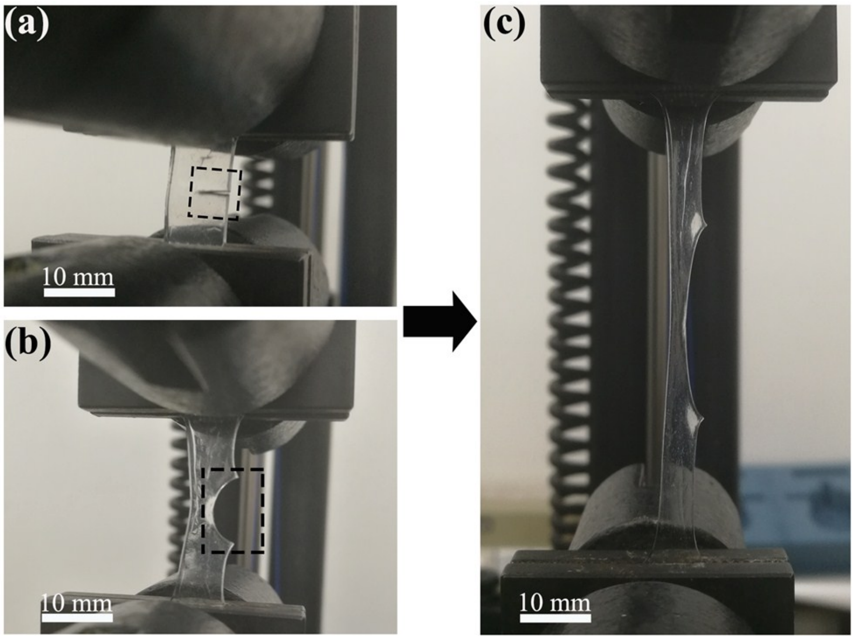

Fig. S10 Images of tensile test of a notch-insensitive and stretchable polymer film. A notch (3.5 mm) was made in DT-IPDI$_2$-HMDI$_1$ polymer film with 1 mm thickness, 15 mm gauge length and 7 mm width and the film were stretched at the loading rate of 100 mm/min. The notch was Passivated and remained stable (right).

Calculation of fracture energies

Fracture tests were conducted using the classical single-edge notch test on the Instron 5567 with a 1KN load cell. A notch of about 3.5 mm in length was made in the middle of a rectangular specimen of about 1.3 mm in thickness and about 7 mm in width. The specimen was fixed in two clamps with a pre-set distance of 15 mm. Then, the specimen was subjected to uniaxial tension with a strain rate of 100 mm/min. The fracture energy (G_c) was calculated by a method which introduces an empirical correction factor of:

$$G_{c=} \; {}^{6Wc}/_{\sqrt{\lambda_c}} \qquad (1)$$

where c is the notch length; W is the strain energy calculated by integration of the stress-strain curve of an un-notched specimen until λ_c; the un-notched specimen underwent a tensioning process with the same strain rate as the notched sample.

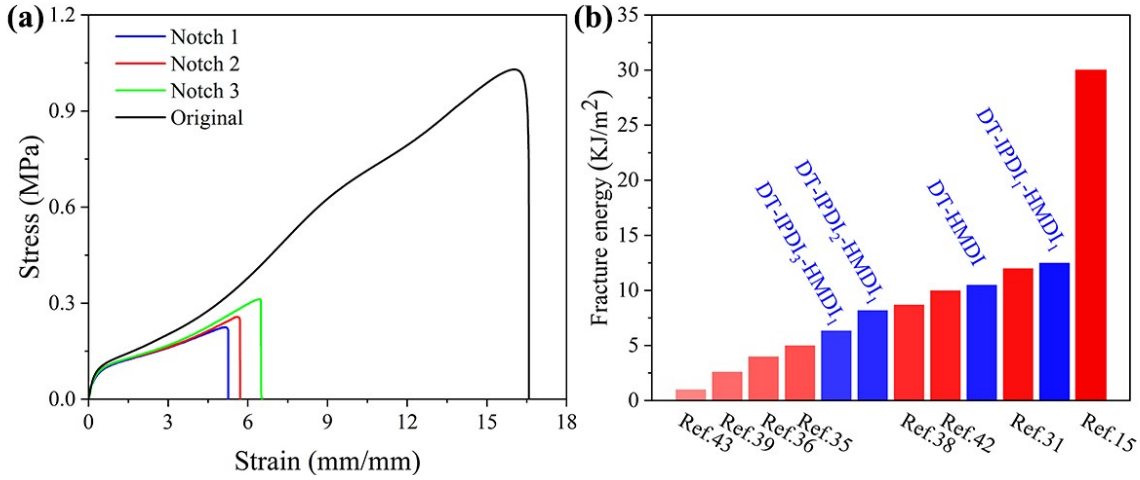

Fig. S11 (a) Stress-extension curves of the unnotched and notched DT-$IPDI_2$-$HMDI_1$ film. (b) Graphic comparison of fracture energies for some classical stretchable polymer elastomers and tough hydrogels.

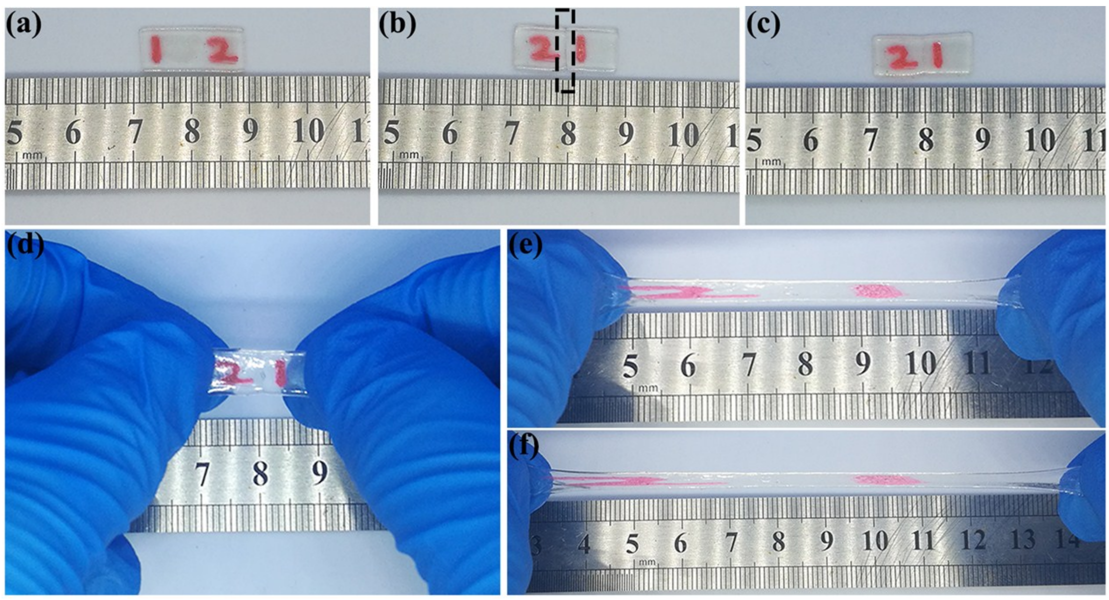

Fig. S12 (a) Images of entire DT-$IPDI_2$-$HMDI_1$ sample was exposed to air for 12h. (b) the film was cut off and linked instead of cut surface. (c) the stitched together film was displaced at room temperature for 6h. d-f) a stretching demonstration was performed at room temperature for 6h after repair of a non-disconnecting surface connection

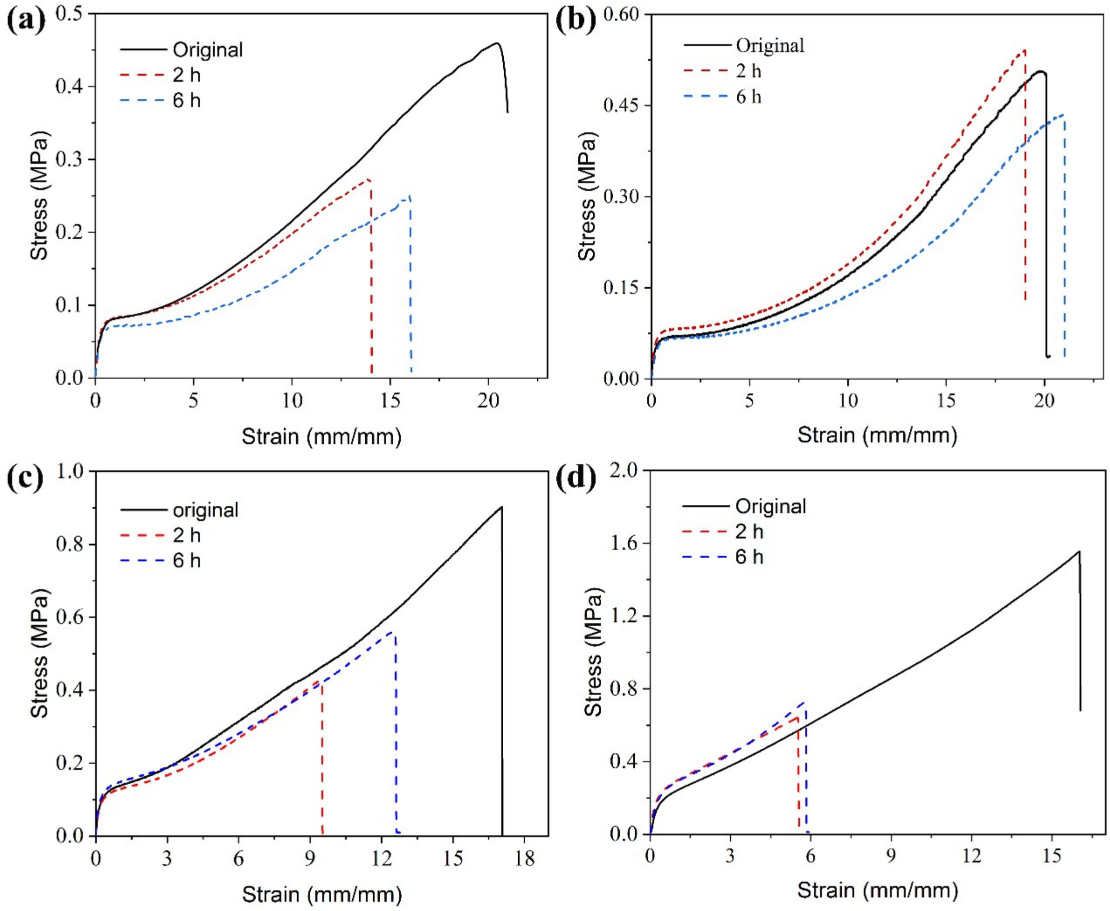

Fig. S13 Stress-strain curves of (a) DT-$IPDI_3$-$HMDI_1$ (b) DT-$IPDI_2$-$HMDI_1$ (c) DT-$IPDI_1$-$HMDI_1$ (d) DT-HMDI were repaired for 2h and 6h, respectively.

Table S1. Summary of the GPC results of prepared elastomers.

Sample Code	M_n (g/mol)	M_w (g/mol)	M_w/M_n
DT-IPDI	13577	15160	1.11
$DT\text{-}IPDI_3\text{-}HMDI_1$	11814	13585	1.14
$DT\text{-}IPDI_2\text{-}HMDI_1$	11784	13732	1.16
$DT\text{-}IPDI_1\text{-}HMDI_1$	11945	13823	1.15
DT-HMDI	12268	15409	1.25

Table S2. The mechanical properties of the samples and the self-repair efficiency of different time were compared

Sample	Young's modulus (MPa)	Breaking strength (MPa)	Elongation at break (%)	Self-healing efficiency (%) after 2h	Self-healing efficiency (%) after 6h	notch-insensitive stretching (%)	Fracture energy (J/m^2)
$DT\text{-}IPDI_3\text{-}HMDI_1$	0.027	0.466	2096.67	66.82	76.45	803.6	5974±367
$DT\text{-}IPDI_2\text{-}HMDI_1$	0.041	0.502	2006.67	94.78	104.55	769.0	7460±800
$DT\text{-}IPDI_1\text{-}HMDI_1$	0.085	0.901	1702.32	55.80	73.72	576.6	11050 ± 1530
DT-HMDI	0.11	1.55	1604.7	34.39	35.89	293.9	9980 ± 698

Table S3. Comparison of the transmittance and the self-healing efficiency of DT-$IPDI_2$-$HMDI_1$ and those reported in related references.

Reference	Substrate	Mechanism	The way of self-healing	Self-healing time（h）	η (%)	T (%)
Ref.16	Polyacrylamide	Host-guest and Hydrogen bond	RT	12	100	80
Ref.23	PDMS	capsule	UV	0.3	100	90
Ref.7	PDMS	Hydrogen bond	RT	1	98.3	80
Ref.25	PU	Hydrogen bond	H_2O	0.05	30	90
Ref.26	PVDF	Ionic ligand	RT	24	100	92
Ref.30	PVA/PDMS	Hydrogen bond	RT	0.25	81	92
Ref.12	bPEI/PAA	Hydrogen bond	H_2O	0.5	100	85
Ref.8	PEM/CNT	Hydrogen bond	H_2O	0.5	100	80.7
Ref.24	Epoxy	Hydrogen bond	140 °C	0.05	100	90
Ref.11	PMMA	capsule	RT	72	100	90
Ref.31	PDMS	Hydrogen bond	RT	48	94	98
Ref.29	PU	2S	70 °C	24	86.3	100
Ref.5	PAA/PDMAPS	Zwitterion +hydrogen bond	RT	48	100	90
Ref.21	Poly-α-lipoic acid	Hydrogen bond	RT	14	86	85
Ref.28	Ionic gel	Metal-ligand and ionic bond	RT	1	100	88.6
Ref.22	PDMS/AgNW-PEDOT	Imine bond	RT	12	86	73
This Work	Polyurea	Hydrogen bond	RT	6	100	92.8

η: the efficiency of self-healing (%), T: Transmittance (%), RT: room temperature