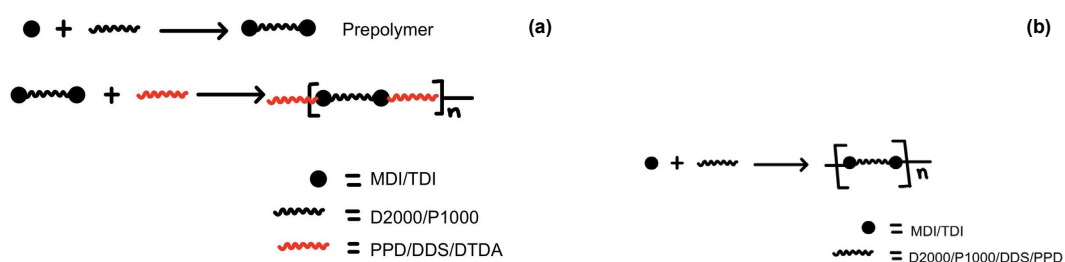


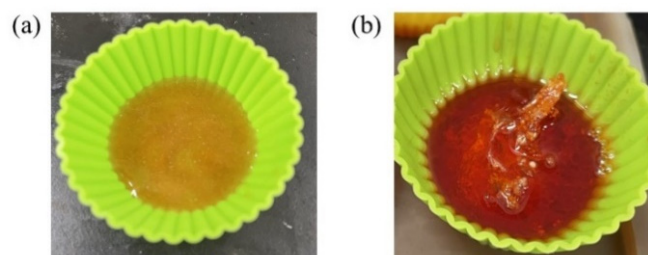
**Table S1.** Names, functions and purpose of materials.

Chemical name	Full name	Function	Purpose
D2000	Poly(propylene glycol) bis (2-aminopropyl ether)	Long chain diamine (soft segment)	
P1000	Polytetramethyleneoxide-di-p-aminobenzoate	Long chain diamine (soft segment)	
IPDI	Isophorone diisocyanate	Aliphatic diisocyanate (hard segment)	IPDI known good self-healing performance but bad mechanical property
MDI	4,4-methylenebis(phenyl isocyanate)	Aromatic diisocyanate (hard segment)	MDI known for poor self-healing performance but good mechanical property
TDI	Toulene 2,4 diisocyanate	Aromatic diisocyanate	Average self-healing performance and average mechanical property
PPD	p-phenylenediamine	Amine chain extender without disulfide bonds	
DDS	Bis(4-aminophenyl) Sulfone	Amine chain extender without disulfide bonds	DDS has bulkier structure than PPD
DTDA	4,4'-dithiodianiline	Amine chain extender with disulfide bonds	Comparison between different chain extenders. DTDA has similar structure as DDS. PPD has lower Mw than DTDA.
THF	Tetrahydrofuran	Solvent	
CHCl <sub>3</sub>	Chloroform	Solvent	

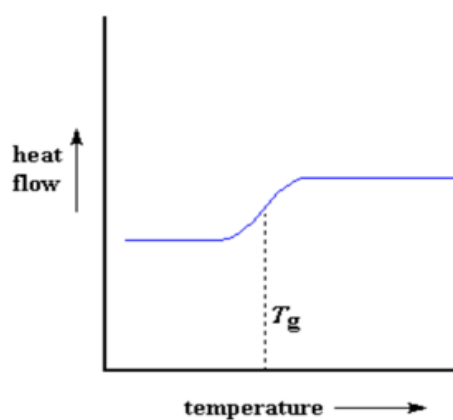
**Table S2.** Initial plan to fabricate polyurea samples.

Sample number	Sample name	Mole of D2000 (mmol)	Mole ratio of IPDI: MDI	Mole of diisocyanate (mmol)	Mole of PPD/DDS/DTDA	Mole ratio of D2000: diisocyanate: chain extender
1	D2000-IPDI-PPD	0.5	1:0	1.0	0.5	1:2:1
2	D2000-IPDI <sub>1</sub> – MDI <sub>1</sub> – PPD	0.5	1:1	1.0	0.5	1:2:1
3	D2000-IPDI <sub>2</sub> – MDI <sub>1</sub> – PPD	0.5	2:1	1.0	0.5	1:2:1
4	D2000-IPDI <sub>3</sub> – MDI <sub>1</sub> –PPD	0.5	3:1	1.0	0.5	1:2:1
5	D2000-MDI-PPD	0.5	0:1	1.0	0.5	1:2:1
6	(Best of 1-5)-DDS	0.5	-	1.0	0.5	1:2:1
7	(Best of 1-5)-DTDA	0.5	-	1.0	0.5	1:2:1

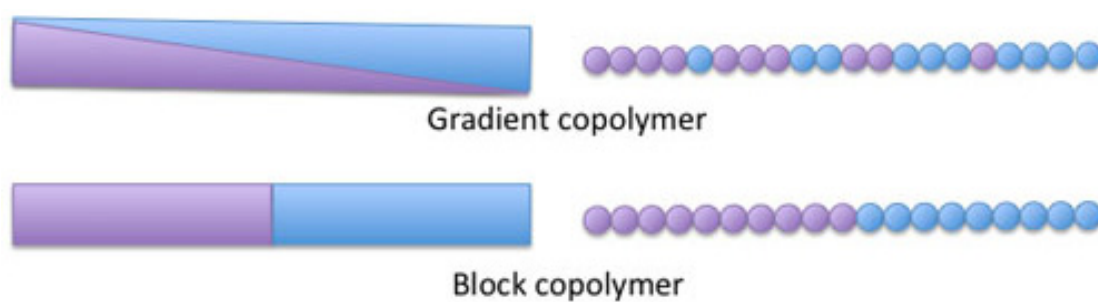
**Figure S1.** Reaction to (a) fabricate polyurea according to initial plan and (b) reaction of diisocyanate with amine.



**Figure S2.** (a) Liquid mixture, and (b) After heating in oven, solid formed and liquid mixture turned red.



**Figure S3.** Glass transition temperature portrayed on DSC graph.



**Figure S4.** Gradient copolymer vs block copolymer.