Supporting Information

Developing self-healable and antibacterial polyacrylate coating with high mechanical strength through crosslinking by multi-amine hyperbranched polysiloxane *via* dynamic vinylogous urethane

Youhao Zhang, Li Yuan, Qingbao Guan, Guozheng Liang* and Aijuan Gu*

State and Local Joint Engineering Laboratory for Novel Functional Polymeric Materials
Jiangsu Key Laboratory of Advanced Functional Polymer Design and Application
Department of Materials Science and Engineering,
College of Chemistry, Chemical Engineering and Materials Science,
Soochow University, Suzhou, 215123, China

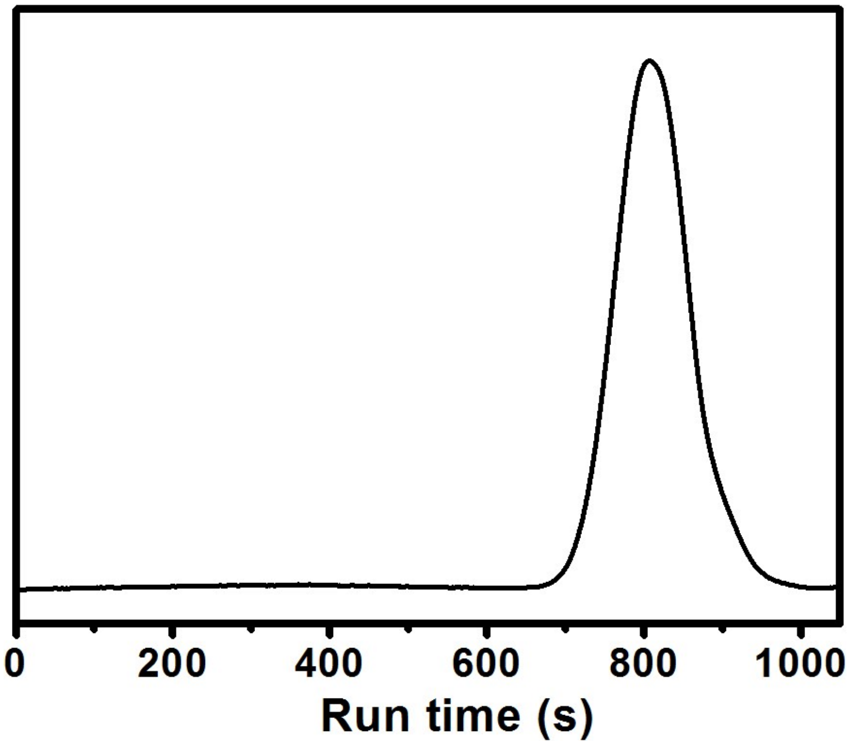

Fig. S1 GPC trace of LP.

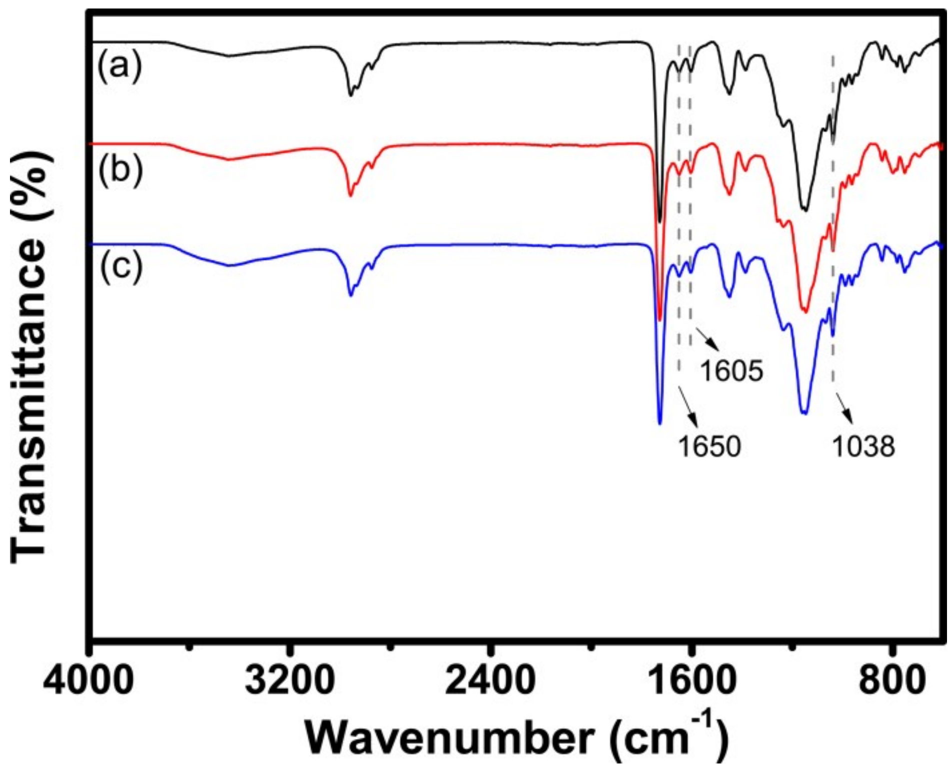

Fig. S2 FTIR spectra of fresh LP-HP6 (a) and those after placed under oxygen atmosphere (b) and immersed in ultrapure water (c).

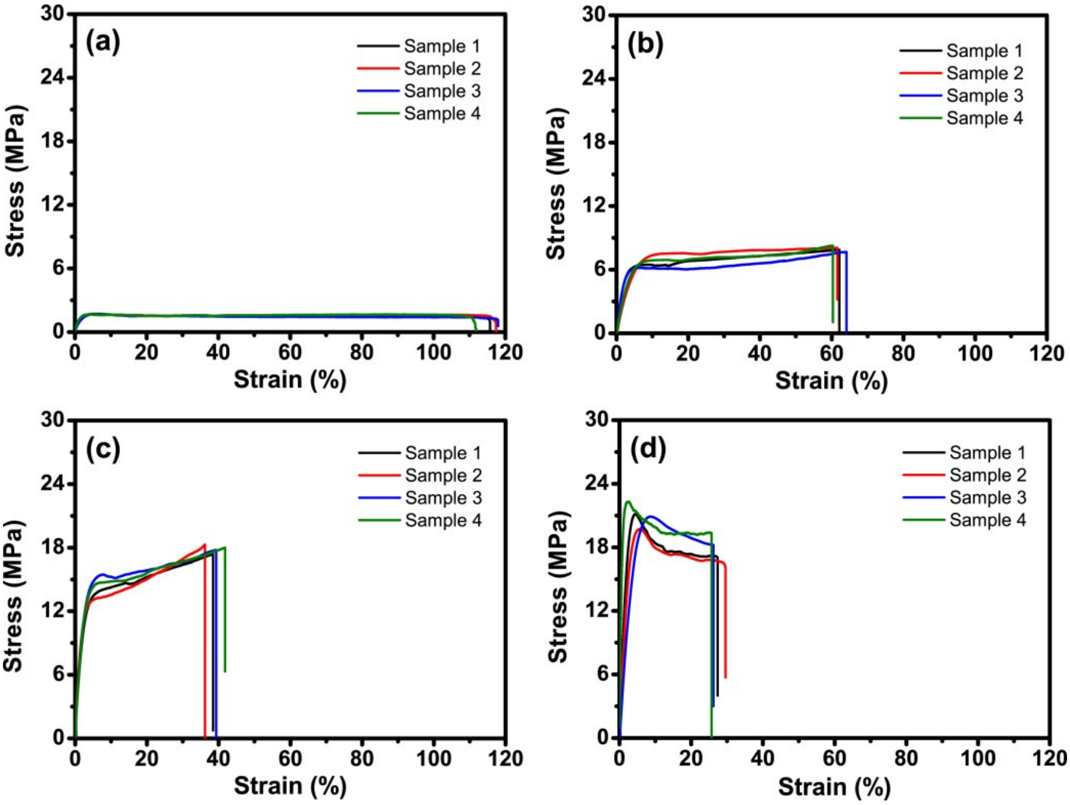

Fig. S3 Tensile stress-strain curves of LP (a) and LP-HP (b: LP-HP3; c: LP-HP6; d: LP-HP9) coatings.

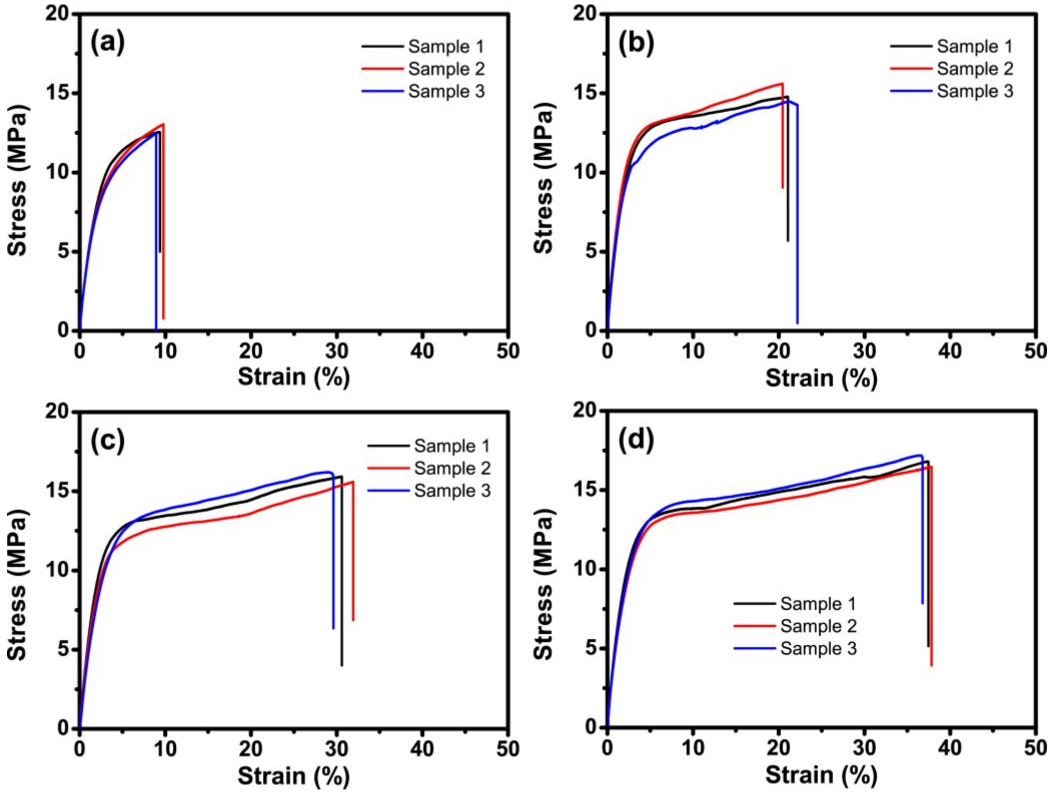

Fig. S4 Tensile stress-strain curves of scratched LP-HP6 coatings with different lengths of self-healing time (a: 0 h; b: 8 h; c: 16 h; d: 24 h).

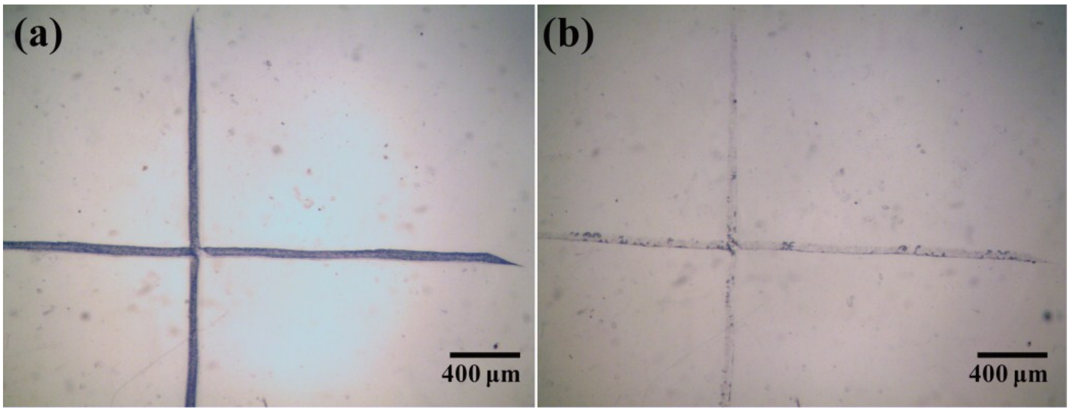

Fig. S5 Optical microscopic images of scratched LP-HP6 coating left in air for 24 h (a) and that after maintained at 60 ^{o}C for 24 h (b).

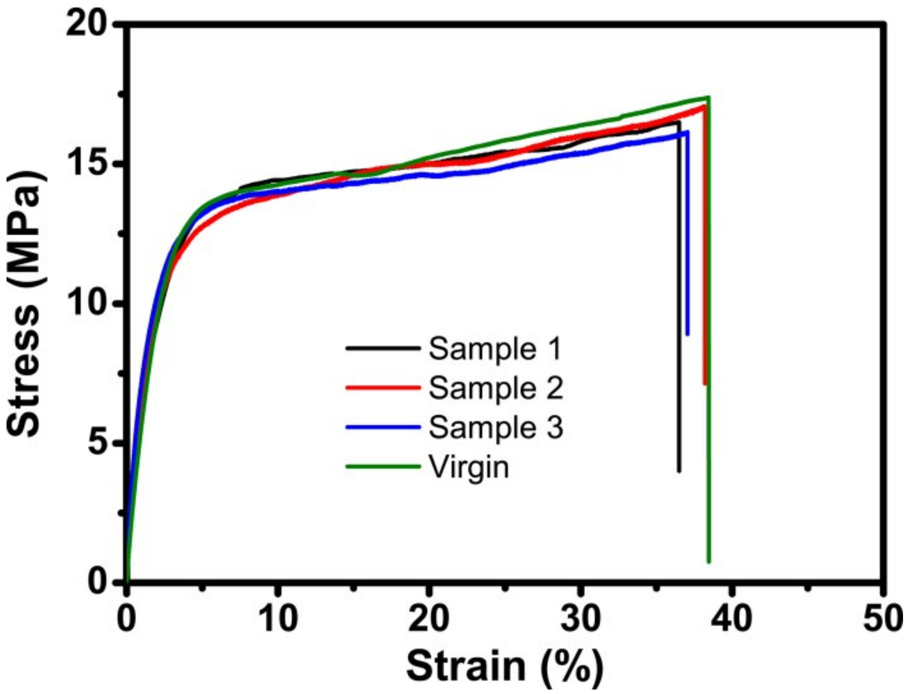

Fig. S6 Tensile stress-strain curves of scratched LP-HP6 coatings after 24 h self-healing (left in air for 24 h before self-healing).

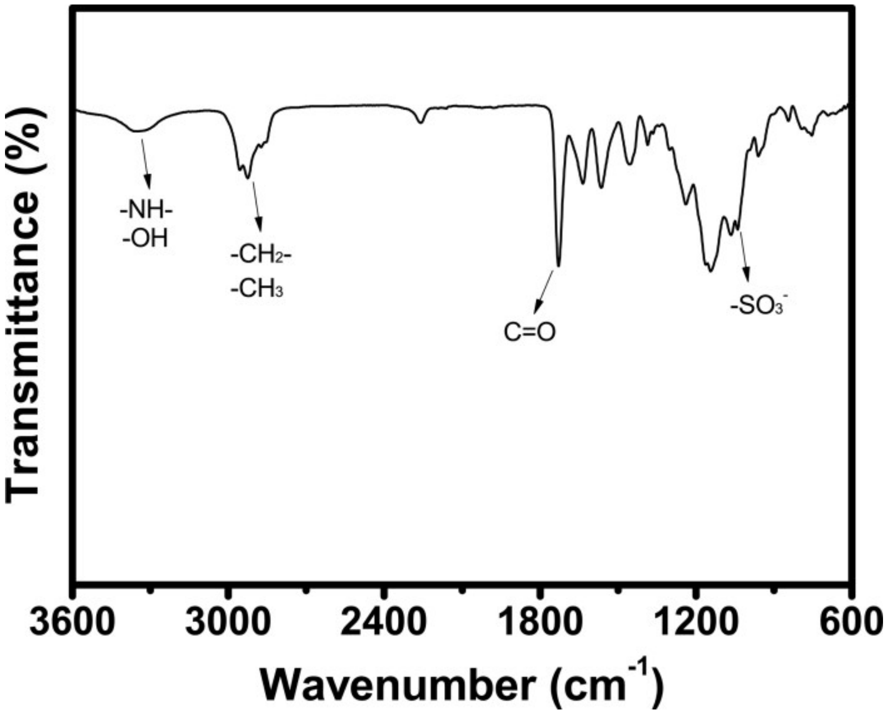

Fig. S7 FTIR spectrum of sLP-HP.

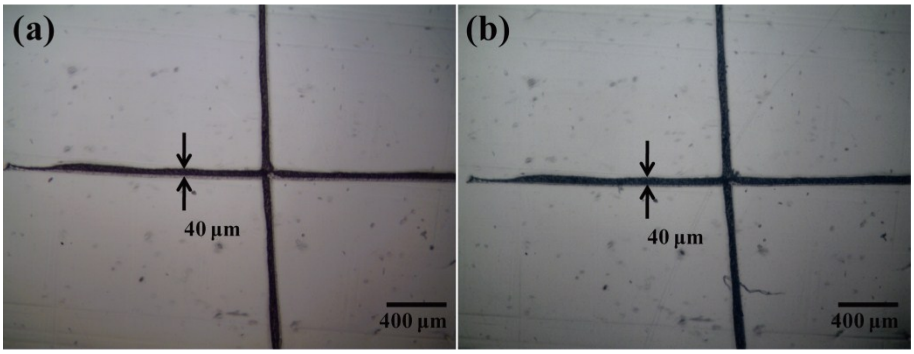

Fig. S8 Optical microscopic images of scratched sLP-HP coating (a) and that after maintained at 60 °C for 24 h (b).

Table S1 Initial degradation temperatures and char yields of LP-HP coatings

Sample	Initial degradation temperature (^{o}C)	Char yield at 800 ^{o}C (wt%)
LP	266	0.58
LP-HP3	246	3.53
LP-HP6	250	6.99
LP-HP9	256	8.09

Table S2 Typical self-healing properties of polyacrylate polymers

Sample name	Self-healing component	T_g (°C)	T_{di} (°C)	Self-healing condition	Self-healing efficiency	Photo[b]	σ_b [c] (MPa)	Ref
MP7	terpyridine and metal ions	64	191	120°C/23h	--[a]	Yes	--	S1
MP9		51	256	90°C/17h	--	Yes	--	
MP11		33	285	100°C/40h	--	Yes	--	
MP12		42	319	60°C/16h	--	Yes	--	
Poly(MMA-co-HEA)	trithioate	--	--	UV/5min	--	Yes	--	S2
MP1	terpyridine and metal ions	85	351	100°C/18h	--	Yes	--	S3
MP3		25	354	80°C/80h	--	Yes	--	
MP4		36	361	100°C/52h	--	Yes	--	
MP9		34	352	90°C/60h	--	Yes	--	
PBA–UPy7.2	2-ureido-4[1 H]-pyrimidinone (UPy)	22	--	R.T./50 h	100 %	--	--	S4
FEF-2-BM	Diels-Alder	-33/90[d]	--	120°C/4h, 25°C/24h	--	Yes	14.2	S5
P2	acylhydrazone	45	303	100°C/64h	--	Yes	--	S6
P3		101	321	150°C/24h	--	Yes	--	
MSP-3	Zn^{2+} and 2,6-bis(1’-methylbenzimidazolyl)pyridine	24/68.4	--	140°C/25min	98%	Yes	4.4	S7
MSP-5		39/79.1	--	140°C/25min	88%	Yes	9.7	
MSP-7		46.8/83	--	140°C/25min	65%	Yes	12.5	
P20/80	hydrogen bonds	33	--	R.T./24h	95%	--	2.9	S8
P20/80-CB5		-	--	R.T./24h	83%	--	4.1	
P20/80-CB20		50	--	R.T./24h	40%	--	7.2	
PHEA-1.5% TMADA	thiol-Michael adduct	9	--	90°C/16h	85%	--	0.22	S9
PHEA-UPy	2-ureido-4[1 H]-pyrimidinone (UPy)	45	200	R.T./2 h, 50% humidity	--	Yes	--	S10
MP2	histidine and zinc salts	53	322	120°C/20h	--		--	S11
MP3		47	295	70°C/38h	--	Yes	--	
MP13		46	296	100°C/20h	--	Yes	--	
MP14		40	297	150°C/40h	--	Yes	--	
MP15		42	240	100°C/20h	--	Yes	--	
LP-HP6	Vinylogous urethane	57	250	60°C/24h	>92%	Yes	17.89 ± 0.42	This work

a. not characterized in the reference.
b. optical microscopic self-healing photos of the sample with Yeses were provided in the reference.
c. value of the virgin sample.
d. T_g values of soft domains and hard domains, respectively.

References

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Table S3 Summary of tensile properties for LP-HP coatings in tensile tests

Coating	Property	Sample 1	Sample 2	Sample 3	Sample 4	Average	Standard deviation
LP	σ_b (MPa)	1.71	1.65	1.67	1.69	1.68	0.03
	ε_b (%)	115.65	117.52	118.32	111.94	115.86	2.84
	E (MPa)	61.54	60.32	63.97	63.97	61.07	2.31
	Toughness (MPa)	1.73	1.84	1.76	1.76	1.75	0.06
LP-HP3	σ_b (MPa)	7.87	8.15	7.69	8.26	7.99	0.26
	ε_b (%)	62.1	60.51	64.83	60.25	61.92	2.10
	E (MPa)	207.66	187.02	215.87	190.32	200.22	13.81
	Toughness (MPa)	4.28	4.5	4.11	4.20	4.27	0.16
LP-HP6	σ_b (MPa)	17.35	18.36	17.84	18.01	17.89	0.42
	ε_b (%)	38.18	36.21	39.51	41.77	38.92	2.34
	E (MPa)	429.43	450.36	425.73	440.70	436.56	11.19
	Toughness (MPa)	5.64	5.33	5.83	6.1	5.72	0.32
LP-HP9	σ_b (MPa)	21.12	19.73	20.95	22.37	21.04	1.08
	ε_b (%)	27.35	29.59	26.14	25.68	27.19	1.75
	E (MPa)	725.09	650.26	635.47	780.77	697.89	67.75
	Toughness (MPa)	4.78	4.96	4.67	5.04	4.86	0.17

Table S4 Summary of tensile properties for each scratched LP-HP6 coatings with different lengths of self-healing time

Coating	Property	Sample 1	Sample 2	Sample 3	Average	Standard deviation
Virgin (0 h)	σ_b (MPa)	12.53	13.07	12.44	12.68	0.34
	ε_b (%)	9.34	9.76	8.89	9.33	0.44
	Toughness (MPa)	0.92	0.96	0.83	0.90	0.07
After 8 h self-healing	σ_b (MPa)	14.77	15.6	14.07	14.81	0.77
	ε_b (%)	21.05	20.11	22.60	21.25	1.26
	Toughness (MPa)	2.68	2.70	2.72	2.70	0.02
After 16 h self-healing	σ_b (MPa)	15.89	15.17	17.12	16.06	0.98
	ε_b (%)	30.53	32.56	28.56	30.55	2.00
	Toughness (MPa)	4.12	4.08	4.03	4.08	0.05
After 24 h self-healing	σ_b (MPa)	16.79	15.89	17.81	16.83	0.96
	ε_b (%)	37.45	38.2	36.23	37.29	0.99
	Toughness (MPa)	5.32	5.2	5.39	5.30	0.09

Table S5 Summary of tensile properties of scratched LP-HP6 coatings after 24 h self-healing (left in air for 24 h before self-healing).

Property	Sample 1	Sample 2	Sample 3	Average	Standard deviation
σ_b (MPa)	16.35	17.03	16.14	16.50	0.46
ε_b (%)	36.21	38.13	37.01	37.11	0.94
Toughness (MPa)	5.19	5.44	5.20	5.27	0.14

Table S6 Self-healing efficiencies of scratched LP-HP6 coatings after 24 h self-healing (left in air for 24 h before self-healing)

Property		Virgin (No scratch)	After 24 h self-healing
σ_b	Value (MPa)	17.89±0.42	16.50±0.46
	Efficiency (%)	-	92.23
ε_b	Value (%)	38.92 ±2.34	37.11±0.94
	Efficiency (%)	-	95.34
Toughness	Value (MPa)	5.72 ±0.32	5.27±0.14
	Efficiency (%)	-	92.13