Supporting information

Fully recyclable high-performance polyacrylsemicarbazide/carbon fiber composites

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Synthesis of ASC model compound (2-benzoyl-N-cyclohexylhydrazine-1-carboxamide): 3 g (3.22 mmol) of benzoylhydrazide and 20 mL of tetrahydrofuran (THF) were added to a 50 mL flask to form a suspension. Then 3.03 g cyclohexane isocyanate (CHI, 24 mmol) was added to the suspension under vigorous stirring. The system became a clear and transparent solution within a few seconds, and then the product was immediately precipitated from the solution. After stirring slowly at room temperature for 12 h, the precipitate was collected by vacuum filtration and the filter cake was washed three times with cold THF. The resulting powder was dried in vacuo at 45 ℃ for 24 h to obtain 4.8 g ASC model compound as a white solid with a yield of ~79.6%. The ^{1}H NMR spectrum of the product is shown in Figure S3a.

^{1}H NMR study of the small model compounds: 2-benzoyl-N-cyclohexylhydrazine-1-carboxamide (0.18 mmol, 0.047 g) and 4-methoxybenzoylhydrazide (0.18 mmol, 0.030 g) were dissolved in 5 mL DMSO-d6. The resulting solution was divided in 7 NMR tubes. The 7 samples were reacted at 120 ℃ for 0, 3, 5, 7, 10, 15 and 30 h respectively, then were measured with ^{1}H NMR spectroscopy.

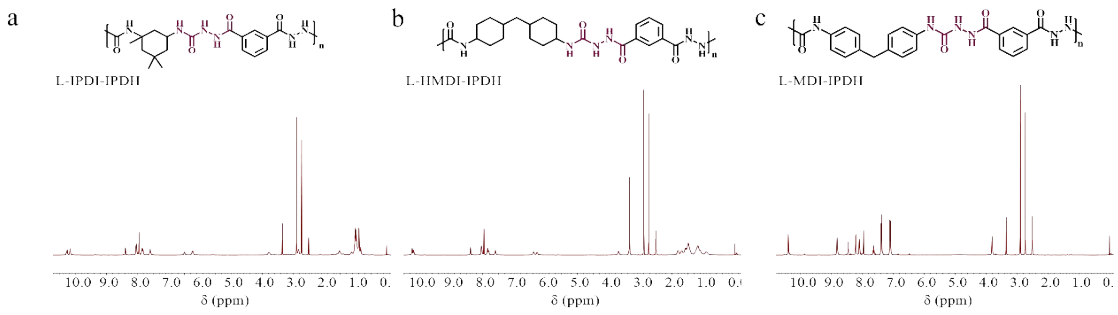

Figure S1 ^{1}H NMR spectra of L-IPDI-IPDH (a), L-HMDI-IPDH (b), L-MDI-IPDH(c).

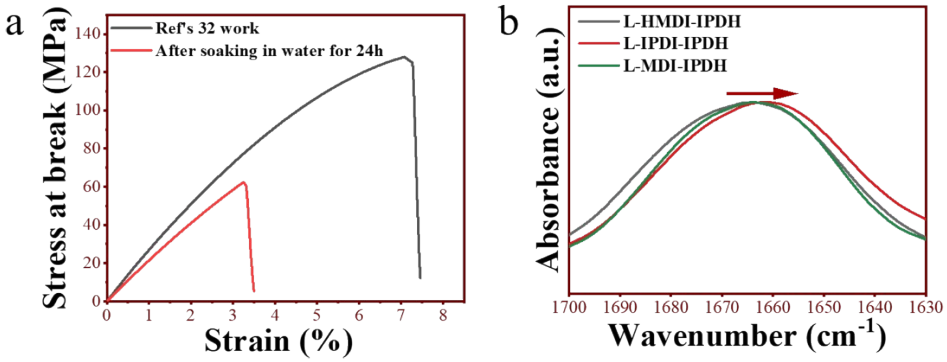

Figure S2 (a) Mechanical properties of PASC-ADH HMDI-0.09 material before and after soaking in water for 24 h at room temperature; (b) The absorption peaks of C=O for L-HMDI-IPDH, L-IPDI-IPDH and L-MDI-IPDH.

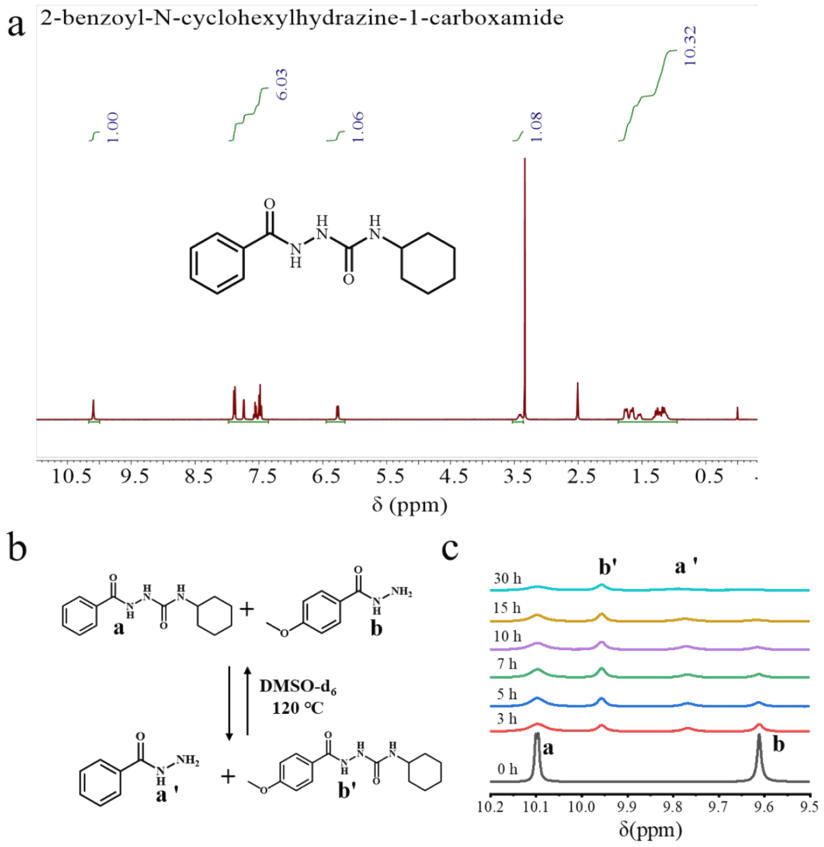

Figure S3 (a) ^{1}H NMR spectrum of 2-benzoyl-N-cyclohexylhydrazine-1-carboxamide (b) schematic illustration of the exchange reaction between model compound 2-benzoyl-N-cyclohexylhydrazine-1-carboxamide and 4-methoxybenzoylhydrazide; (c) ^{1}H NMR spectra as a function of time.

Figure S4 The optical pictures showing the dimensional stability of PASC materials with different crosslinking index in conventional solvents.

Table S1 Mechanical properties of L-IPDI-IPDH, L-HMDI-IPDH, L-MDI-IPDH, PASC-ADH-HMDI-0.09 and the Mn measured by GPC

Sample	Initial modulus (GPa)	Stress at break (MPa)	Strain at break (%)	Mn
L-IPDI-IPDH	3.3±0.2	71.1±14.1	2.7±0.5	12962
L-IPDI-IPDH ^{wet}	3.3±0.2	65.6±8.5	2.04±0.1	
（RT，30 d)	(102.8 %)	(92.3 %）	（75.6%）	
L-IPDI-IPDH ^{wet}	2.1±0.2	31.5±4.3	2.2±0.3	
（60 ℃，7 d)	(62.0%)	(44.3%）	（82.9%）	
L-HMDI-IPDH	2.4±0.2	97.3±11.6	11.3±0.6	11136
L-HMDI-IPDH ^{wet}	2.5±0.3	81.8±6.3	14.8±3.7	
（RT，30 d)	（108.5%）	（84.1%）	（131.5%）	
L-HMDI-IPDH ^{wet}	1.1±0.2	33.8±5.4	11.7±4.3	
（60 ℃，7 d)	（45.3%）	（34.7%）	（103.9%）	
L-MDI-IPDH	2.3±0.4	102.6±1.7	5.9±1.6	16557
L-MDI-IPDH ^{wet}	2.7±0.2	92.6±2.9	8.3±0.9	
（RT，30 d)	（115.3 %）	（90.3%）	（39.7%）	
L-MDI-IPDH ^{wet}	1.3±0.2	24.9±5.1	2.4±0.3	
（60 ℃，7 d)	（54.0%）	（24.3%）	（39.7%）	
PASC-ADH-HMDI-0.09	2.4±0.4	110±2.7	8.1±0.8	
PASC-ADH-HMDI-0.09^{wet}	1.8±0.2	62±1.2	3.3±0.9	
(RT,24 h)	(75%)	(56%)	(41%)	

Table S2 Summary of the assignment of the deconvoluted subpeaks in the FTIR C=O absorption bands for the L-IPDI-IPDH, L-HMDI-IPDH and L-MDI-IPDH.

	L-IPDI-IPDH		**L-HMDI-IPDH**		**L-MDI-IPDH**	
	Peak area	Peak position	Peak area	Peak position	Peak area	Peak position
Free C=O	1.2 ± 0.6	1710.1 ± 1.8	2.8 ± 0.3	1721.1± 0.3	3.5± 0.9	1719.2 ± 2.1
Disordered H-bonded	10.3 ± 2.6	1679.1 ± 0.9	28.4 ± 1.8	1677.1 ± 0.3	24.3 ± 2.4	1675.4 ± 0.9
Ordered H-bonded	45.1 ± 2.7	1659.6± 0.8	37.9 ± 1.8	1657.9± 0.3	27.6 ± 2.8	1657.3± 0.7
the fractions of H-bonded	97.9 %		95.9 %		93.6 %	
Proportion of ordered hydrogen bonds	81.5 %		57.2 %		53.2 %	

Table S3 Mechanical properties of carbon fiber prepreg with different resin content

Sample	**Initial modulus (GPa)**	**Stress at break (MPa)**	**Strain at break (%)**
CFRP-60% prepreg	36.8±3.1	472.6±25.6	1.8±0.2
CFRP-50% prepreg	37.7±4.9	378.1±38.4	1.5±0.1
CFRP-40% prepreg	20.1±1.8	231.2±25.3	2.5±1.3

Table S4 Flexural properties of carbon fiber double-layer laminates with different resin content

Sample	**Flexural modulus (GPa)**	**Flexural stress (MPa)**	**Flexural strain (%)**
2-CFRP-60%	27.7±5.7	157.3±7.3	0.8±0.3
2-CFRP-50%	29.8±2.1	326.9±37.4	2.01±0.4
2-CFRP-40%	12.7±1.5	243.8±7.4	2.96±0.2