

Supporting Information

A kind of self-healing and recyclable poly(urea-imine) thermoset synthesized from CO₂

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Measurement of molecular weight of oligourea

The M_n of CO₂-based oligourea (OUa) was measured by acid-base titration (ASTM D2074-92) with a two-electrode automatic potentiometric titrator. The 0.1 M HCl/isopropanol standard solution was used as the titrate. The M_n was calculated by the following formula. The results were recorded using the mean number of three tests.

$$Mn = \frac{m}{c \times V/N} \times 1000$$

Where Mn was the number average molecular weight of the CO₂-oligourea, g/mol; c was the concentration of HCl/isopropanol solution, mol/L; V was the Volume of HCl/isopropanol solution used at titration end point, ml; m was the mass of the CO₂-oligourea used, g; N was the number of the end group for CO₂-oligourea, N is 2. As a result, M_n of the CO₂-based oligourea (OUa) used was 890 g mol⁻¹.

Recovery by chemical methods

The OUa₂-T₃ sample (6.85 g) can be completely dissolved in methanol (100 mL) to form a homogenous yellow solution with adding new OUa (4.8 mmol). By adding new TA (15.6 mmol) and TAEA (7.2 mmol), the solution was stirred at 25 °C for 4 h and then placed in the Teflon mould. Then methanol was evaporated at room temperature for 24 h and the sample was dried at 80 °C for 12 h. A recycled OUa₂-T₃ sample was obtained.

Degradation process

OUa₂-T₃ was cut into rectangular samples and then immersed respectively in a 10 mL of methanol/H₂O (V:V = 1:1) and 10 mL 0.1 M HCl of methanol/H₂O (V:V = 1:1) at room temperature. The degradation rate was calculated by weighing method, and the obtained degraded mixture was analyzed by FT-IR and ¹³C NMR after evaporating out solvent.

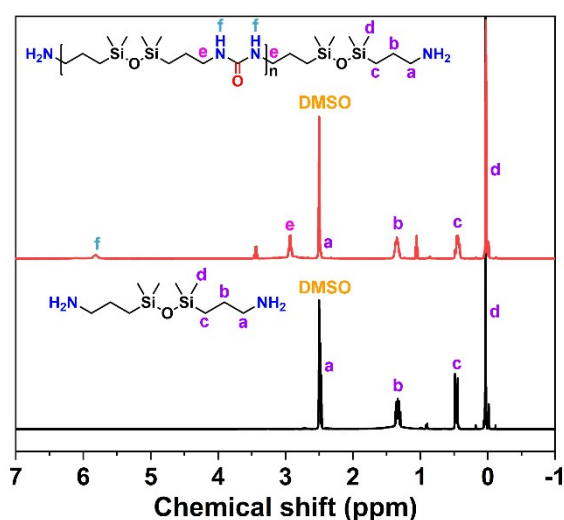


Fig. S1 ¹H NMR spectra of 1,3-bis(3-aminopropyl)tetramethyldisiloxane and oligourea.

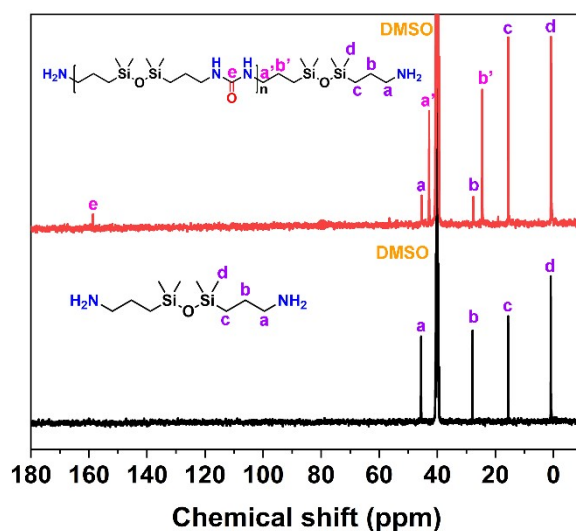


Fig. S2 ^{13}C NMR spectra of 1,3-bis(3-aminopropyl)tetramethyldisiloxane and oligourea.

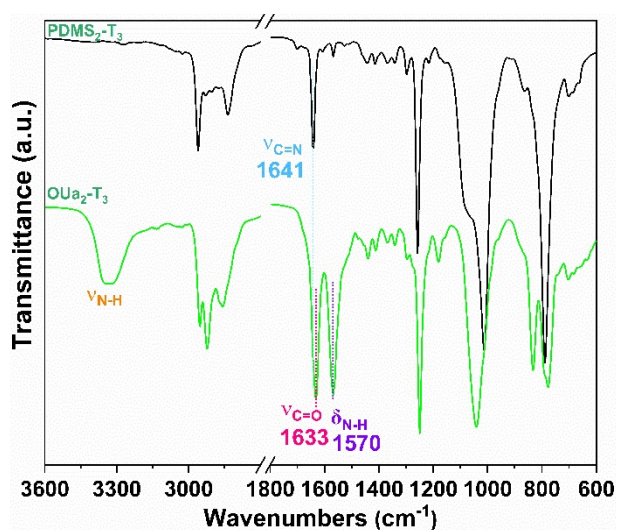


Fig. S3 FT-IR of PDMS₂-T₃ and OUa₂-T₃.

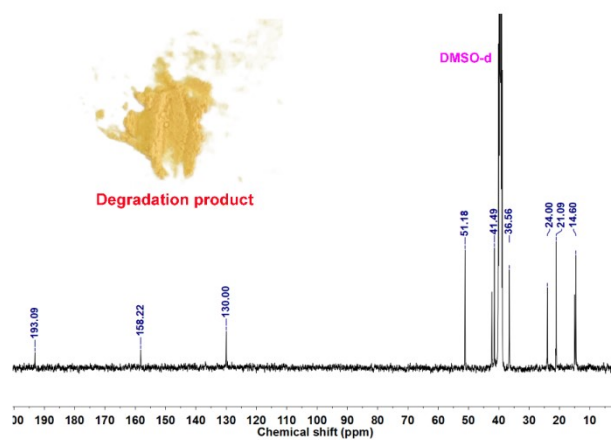


Fig. S4 ^{13}C NMR spectra of the degradation product of OUa₂-T₃ in 0.1 M HCl methanol/H₂O.

Table S1 Summary of the OUa_x-T_y samples and their mechanical and thermal properties.

Sample	OUa, TREA, TA molar ratio	T _g (°C) ^a	T _{d,5%} (°C) ^b	Tensile stress (MPa)	Elongation at break (%)	Young's modulus (MPa)
OUa ₁ -T ₀	1:0:1	-7	275	/	/	/
OUa ₃ -T ₂	3:2:6	4	267	1.3 ± 0.1	257 ± 18	3.3 ± 0.5
OUa ₁ -T ₁	1:1:2.5	6	268	4.3 ± 0.5	98 ± 10	56 ± 8
OUa ₂ -T ₃	2:3:6.5	11	274	10.2 ± 0.2	51 ± 6	79 ± 8
OUa ₀ -T ₁	0:1:1.5	123	250	23.5 ± 2.6	4 ± 0.3	620 ± 45

^a T_g is glass transition temperature, which is measured by DSC.

^b T_{d,5%} is the initial decomposition temperature (the temperature of 5% weight loss).

Table S2 Mechanical properties of original and self-healed OUa₂-T₃.

Entry	Tensile stress (MPa)	Elongation at break (%)	Young's modulus (MPa)
Original	10.2 ± 0.2	51 ± 6	79 ± 8
Healed	9.4 ± 0.3	53 ± 4	63 ± 6

Table S3 Mechanical properties of original and reprocessed OUa₂-T₃.

Entry	Tensile stress (MPa)	Elongation at break (%)	Young's modulus (MPa)
Original	10.2 ± 0.2	51 ± 6	79 ± 8
1st reprocessed	10.6 ± 0.1	54 ± 3	74 ± 4
2nd reprocessed	9.7 ± 0.8	52 ± 2	72 ± 10
3rd reprocessed	10.3 ± 0.6	52 ± 3	78 ± 3

Table S4 Mechanical properties of original and reprocessed OUa₂-T₃.

Entry	Tensile stress (MPa)	Elongation at break (%)	Young's modulus (MPa)
Original	10.2 ± 0.2	51 ± 6	79 ± 8
Recycled	7.9 ± 1.5	54 ± 1	67 ± 9