Electronic Supplementary Information for

Design of A Mechanically Strong and Highly Stretchable Thermoplastic Silicone Elastomer Based on Coulombic Interactions

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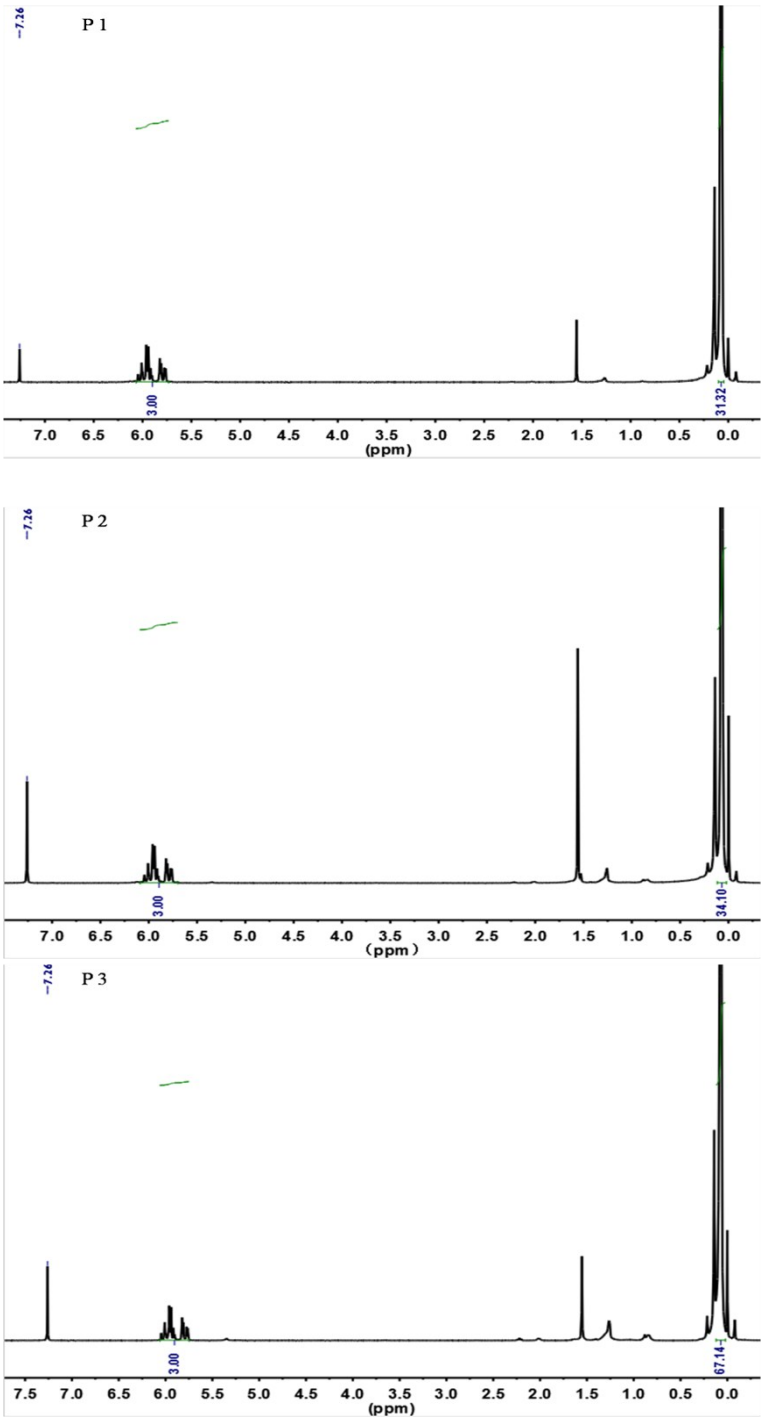

Fig. S1 ^{1}H NMR spectra of the P1, P2 and P3 in $CDCl_3$.

Mole fraction of vinyl groups in polymer was calculated as:

$\frac{I_{Vi}/3}{(I_{Vi}/3)+(I_{Me}/6)}$, I_{Vi} is the integral of vinyl protons at δ = 5.5 – 6.3 ppm, I_{Me} is the integral of methyl protons at δ = 0 – 0.2 ppm.

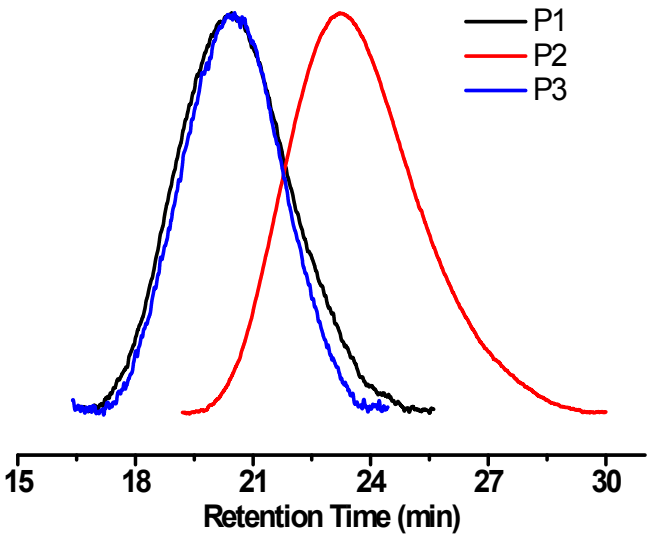

Fig. S2 GPC results of PDMS precursors.

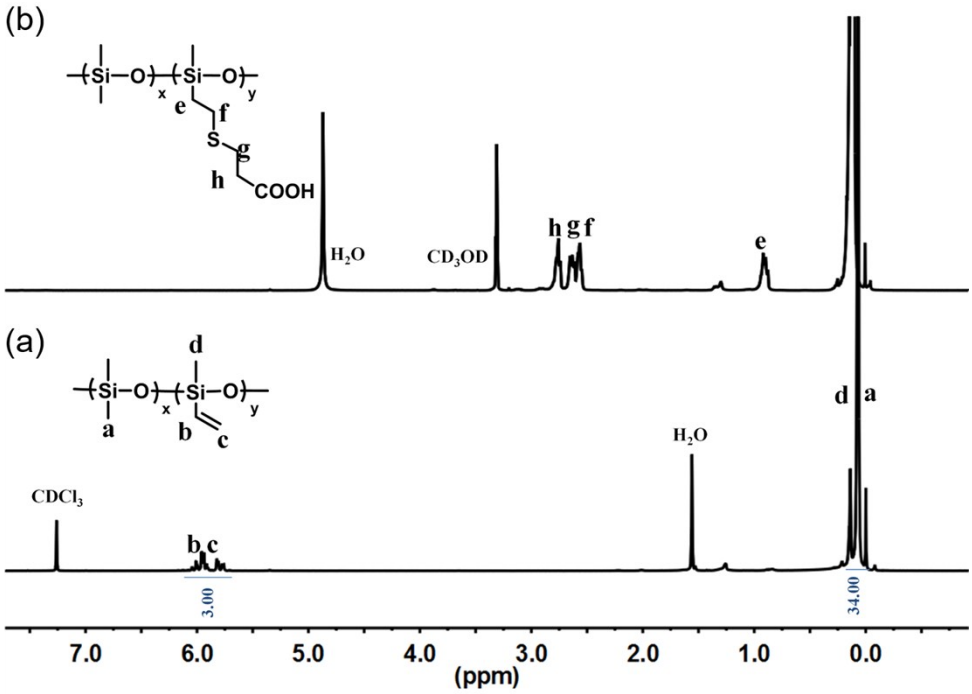

Fig. S3 ^{1}H NMR spectra of the (a) P2 in $CDCl_3$ and (b) P2-*g*-COOH in CD_3OD.

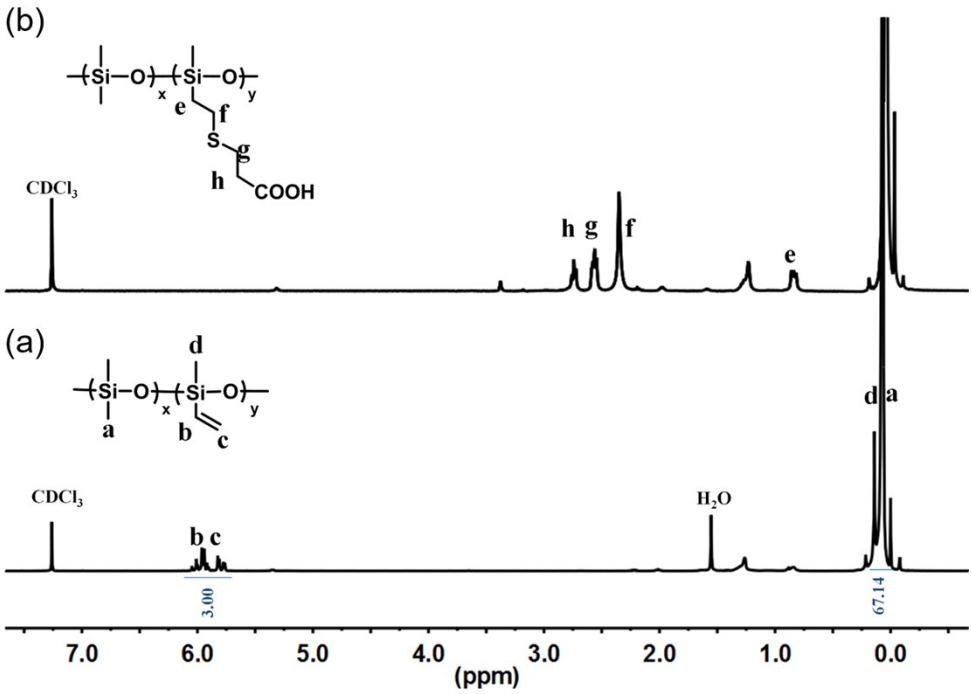

Fig. S4 ^{1}H NMR spectra of (a) P3 in $CDCl_3$ and (b) P3-*g*-COOH in $CDCl_3$.

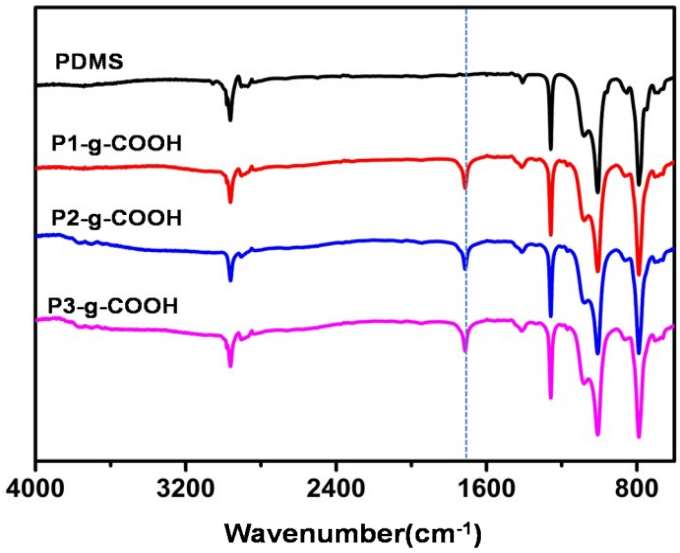

Fig. S5 FTIR spectra of P1 PDMS, P1-*g*-COOH, P2-*g*-COOH, P3-*g*-COOH.

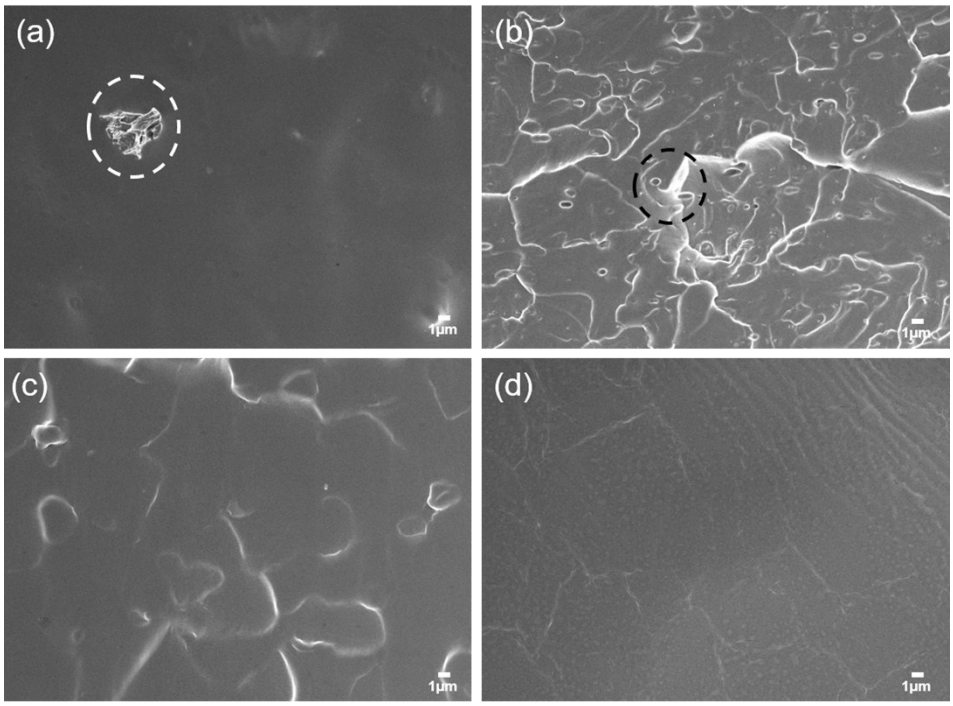

Fig. S6 SEM images of the fractured surfaces of P1-*g*-COOH/ZnO elastomers: (a, b) COOH/ZnO=1/1 and (c, d) COOH/ZnO=2/1, (a, c) before and (b, d) after treated with 2.5 M hydrochloric acid solution for 5 min. (The scanning electron microscopy (SEM) images were obtained on a JSM-7500F SEM (from JEOL, Japan) at 5 kV. The fractured surfaces of the obtained elastomers were coated by a thin layer of gold before scanning.)

Table S1 Tensile stress−strain results of PDMS-*g*-COOH/ZnO composites with molar ratio of COOH/ZnO = 1.5/1 at the stretching speed of 10 mm/min

PDMS precursor	Breaking strength (MPa)	Elongation at break (%)
P1	5.68	637
P2	4.11	735
P3	4.62	756

Table S2 Tensile stress−strain results of P1-*g*-COOH/ZnO composite with different molar ratios of COOH/ZnO at the stretching speed of 10 mm/min

COOH/ZnO	Breaking strength (MPa)	Elongation at break (%)
2/1	3.08	876
1.5/1	5.68	637
1/1	6.47	523

Table S3 Tensile stress−strain results of P1-*g*-COOH/ZnO composite with the molar ratio of COOH/ZnO = 2/1 at different stretching speeds

Stretching speed (mm/min)	Breaking strength (MPa)	Elongation at break (%)

5	2.04	1300
10	3.08	876
50	5.13	660
100	5.47	483

Table S4 Self-healing efficiencies of P1-*g*-COOH/ZnO composite with the molar ratio of COOH/ZnO = 1.5/1 at different T for 4 h

T (°C)	healing efficiency of strain (%)	healing efficiency of strength (%)
30	9.3	51.1
50	55.3	73.9
80	107.7	83.5

Table S5 Self-healing efficiencies of P1-*g*-COOH/ZnO composite with the molar ratio of COOH/ZnO = 1.5/1 at 80 °C after different durations

t (h)	healing efficiency of strain (%)	healing efficiency of strength (%)
0.5	35.6	57.0
1	46.5	59.0
2	77.7	77.6
4	107.7	83.5