Supporting Information

High-strength Fluorosilicone Rubber with Exceptional Shape Memory Performance through Stereochemical Structure Regulation

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Table S1 Sample information and experimental formulations for PMTFPS.

Number	D_3F (cis-D_3F/trans-D_3F) (g)	D_4 ^{vi} (g)	Alkali catalysts (g)	M_η ($\times10^4$g/mol)	Vi (%mol)
1	200(0/100)	0.39	0.62	64	0.33
2	200(0/100)	0.38	0.55	80	0.32
3	200(0/100)	0.37	0.50	85	0.33
4	200(0/100)	0.36	0.43	99	0.33
5	200(0/100)	0.38	0.40	111	0.34
6	200(0/100)	0.38	0.35	130	0.33
7	200(40/60)	0.38	0.60	55.6	0.32
8	200(40/60)	0.37	0.51	85	0.33
9	200(40/60)	0.38	0.39	119	0.32
10	200(40/60)	0.39	0.34	130	0.33
11	200(70/30)	0.38	0.62	61	0.32
12	200(70/30)	0.39	0.58	80	0.34
13	200(70/30)	0.15	0.50	85	0.12
14	200(70/30)	0.22	0.51	85	0.17
15	200(70/30)	0.38	0.52	85	0.33
16	200(70/30)	0.36	0.42	100	0.34
17	200(70/30)	0.38	0.38	118	0.32

Note: 0.3 g promoter was added to all experiments; the promoter only accelerated the reaction rate but did not affect M_η and Vi content.

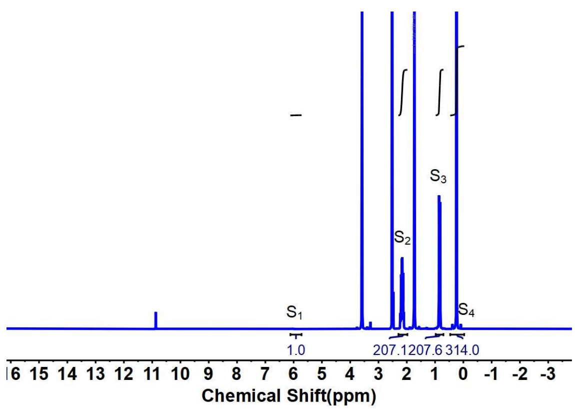

Figure S1 ^{1}H NMR spectra of PMTFPS-70.

[1]H NMR of PMTFPS was tested on a Bruker AVANCE III HD400 spectrometer with tetrahydrofuran-d_8 as the solvent. And the sample was scanned 128 times. In Figure S1: peak 1 (δ=5.80-6.00 ppm) corresponds to - C***H***=C***H₂***; Peak 2 (δ=2.13-2.16 ppm) and peak 3 (δ=0.85-0.89 ppm) correspond to - $CH_2C\boldsymbol{H_2}CF_3$ and - $C\boldsymbol{H_2}CH_2CF_3$, respectively; Peak 4 (δ=0.07 ppm) corresponds to - $C\boldsymbol{H_3}$. Therefore, the actual Vi content was calculated using the Equation S1:

$$Vi(\%mol) = \frac{S_1/3}{S_1/3 + S_4/3} = \frac{S_1}{S_1 + S_4} \quad \text{(S1)}$$

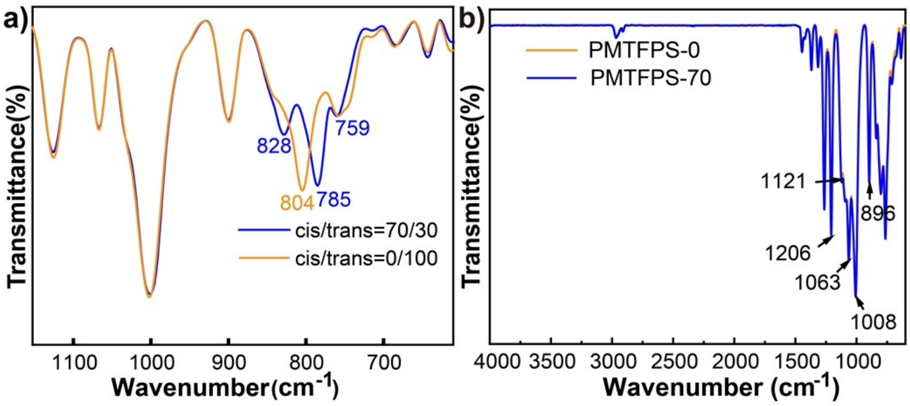

Figure S2 (a) Local FT-IR spectra of D_3F. (b) FT-IR spectra of PMTFPS.

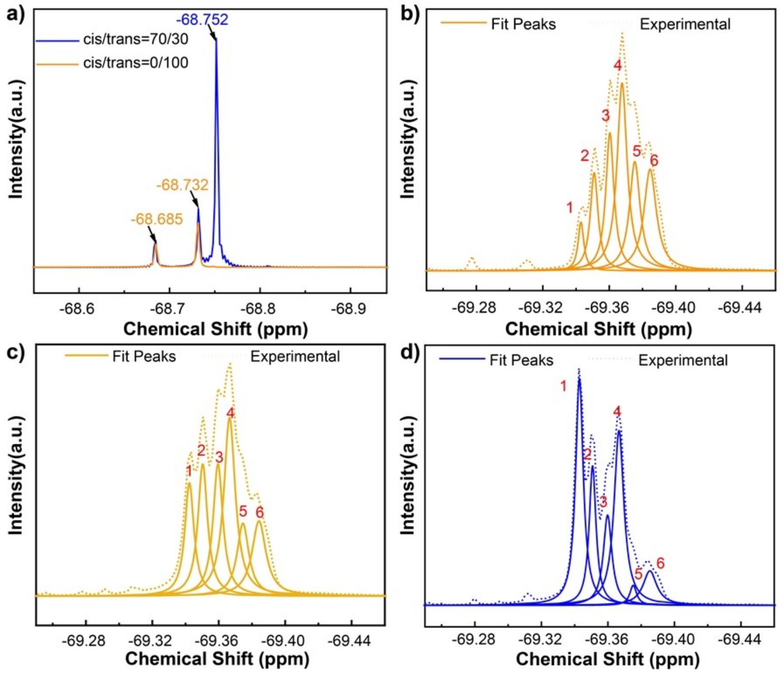

Figure S3 (a) ^{19}F NMR spectra of D_3F. Fitting curves of ^{19}F NMR: (b) PMTFPS-0; (c) PMTFPS-40 and (d) PMTFPS-70.

Table S2 The chemical shift and area corresponding to the fitting peak of ^{19}F NMR（PMTFPS-0）.

Number	1 (mmm)	2 (rmm/mmr)	3 (mrm)	4 (rmr)	5 (rrm/mrr)	6 (rrr)
Chemical Shift (ppm)	-69.343	-69.350	-69.360	-69.366	-69.375	-69.385
Area (%)	3.3	8.7	23.4	28.9	18.0	17.7

Table S3 The chemical shift and area corresponding to the fitting peak of ^{19}F NMR（PMTFPS-40）.

Number	1 (mmm)	2 (rmm/mmr)	3 (mrm)	4 (rmr)	5 (rrm/mrr)	6 (rrr)
Chemical Shift (ppm)	-69.343	-69.350	-69.360	-69.366	-69.375	-69.385
Area (%)	13.6	16.8	17.2	27.1	11.4	13.9

Table S4 The chemical shift and area corresponding to the fitting peak of ^{19}F NMR（PMTFPS-70）.

Number	1 (mmm)	2 (rmm/mmr)	3 (mrm)	4 (rmr)	5 (rrm/mrr)	6 (rrr)
Chemical Shift (ppm)	-69.343	-69.350	-69.360	-69.366	-69.375	-69.385
Area (%)	29.1	16.5	13.3	29.6	3.4	8.1

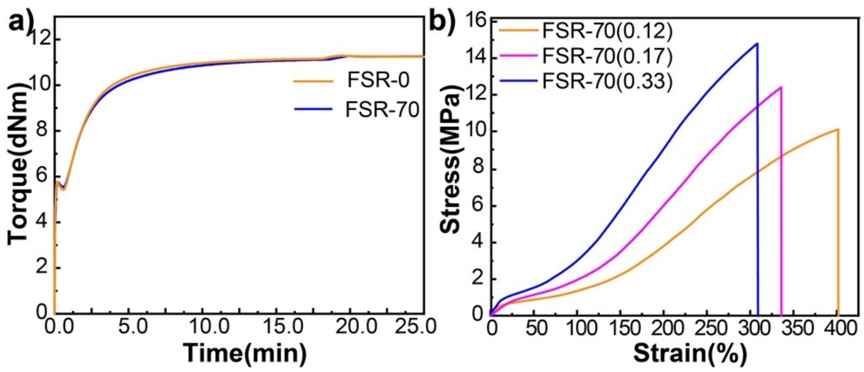

Figure S4 (a)The vulcanization curves of FSR. (b)The stress-strain curves of FSR-70 with Vi=0.12, 0.17, 0.33 %mol.

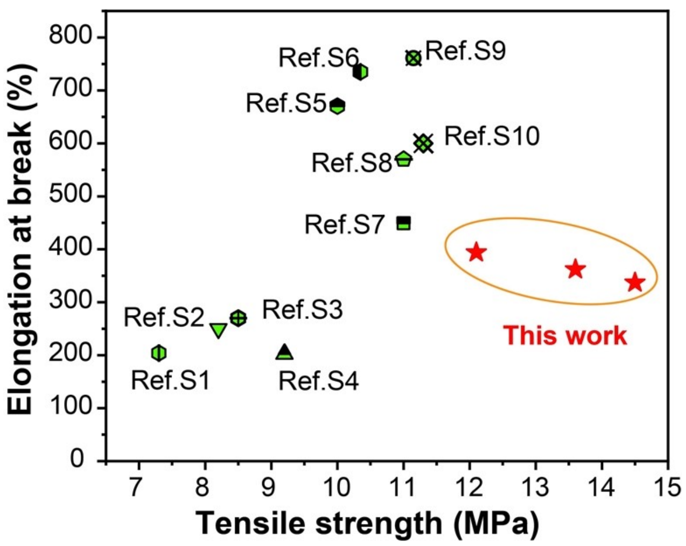

Figure S5 Comparison of our work with recent works in FSR elastomer[1-10]

Table S5 Crosslinking densities of FSR-0.

M_η(×10^4 g/mol)	64	80	85	99	111	130
Crosslinking density (× 10^{-4} mol/cm^3)	1.43	1.41	1.45	1.46	1.48	1.47

Table S6 Crosslinking densities of FSR-40.

M_η(×10^4 g/mol)	85
Crosslinking density (× 10^{-4} mol/cm^3)	1.40

Table S7 Crosslinking densities of FSR-70.

M_η(×10^4 g/mol)	61	80	85	100	118
Crosslinking density (× 10^{-4} mol/cm^3)	1.44	1.42	1.50	1.47	1.45

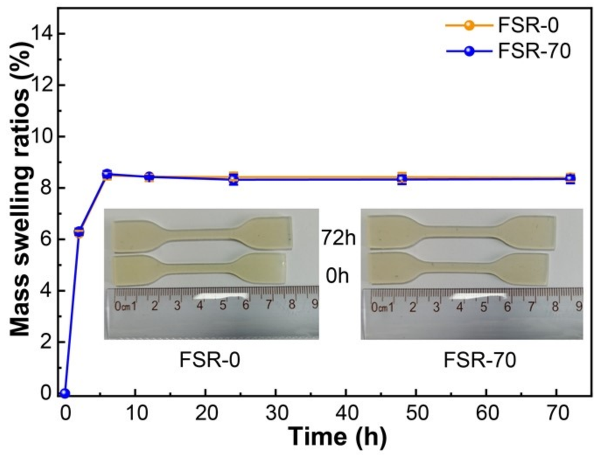

Figure S6 The trend of mass swelling rate of FSR with soaking time and digital photos of FSR before and after soaking.

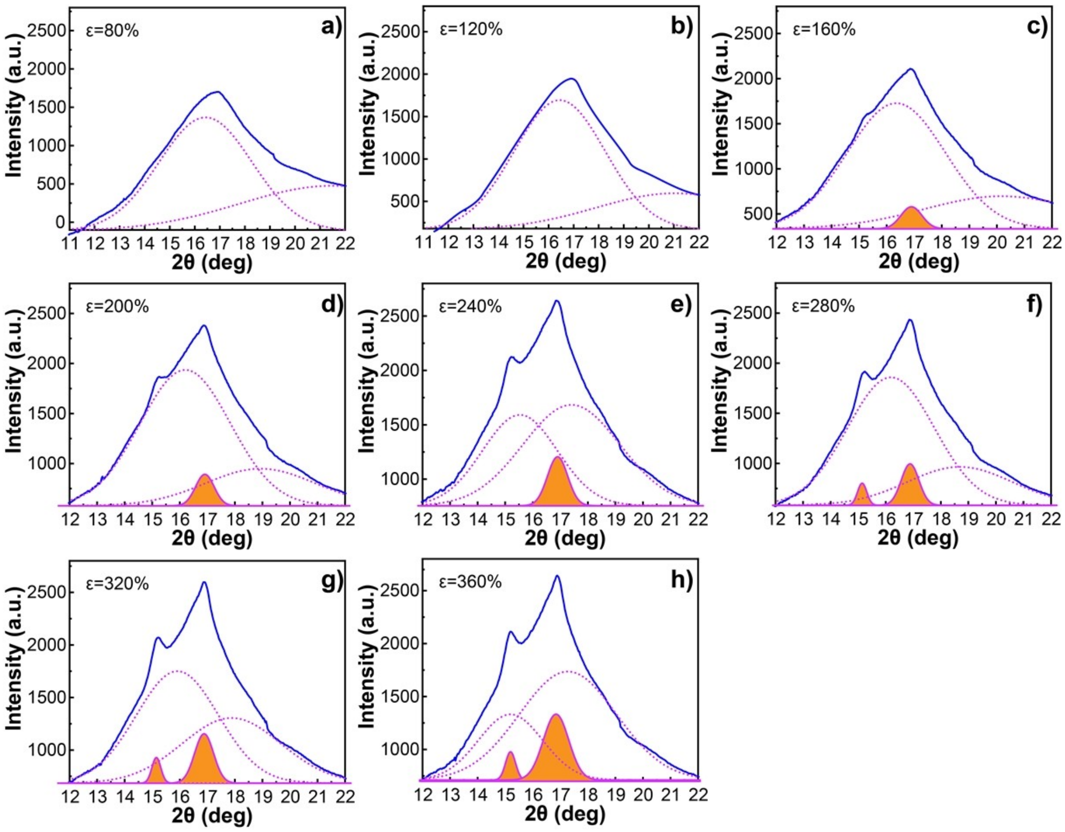

Figure S7 Fitting curves of 1D-WAXS with different strains (Dashed line: amorphous peaks; Shaded: crystalline peaks).

Table S8 The shape fixation rate (R_f) and the shape recovery rate (R_r) of FSR-70.

	1st cycle	2st cycle	3st cycle	4st cycle	5st cycle
R_f(%)	99.2	99.2	99.7	99.2	99.2
Rr (%)	94.5	99.4	100	100	99.5

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