Supporting Information

Thermoplastic Polyurethane/Silicone Rubber (TPU/SiR) Thermoplastic Elastomers with Fine Phase Structure and Comfortable Texture based on Polymerization-induced Phase Separation

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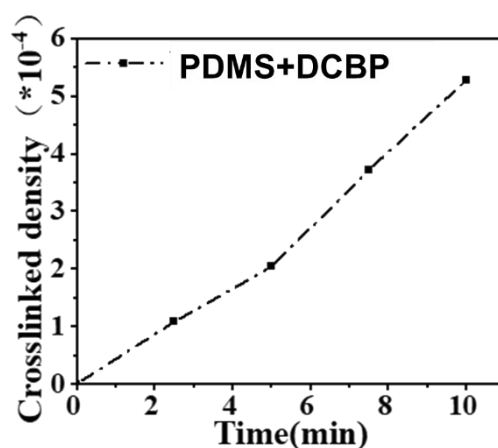[*]Corresponding author: tianm@mail.buct.edu.cn; yubing@mail.buct.edu.cn

Experimental Section

Table S1. The formulation for preparation of TPU/SiR TPEs with different pre-polymerization time

Sample	Formula					pre-polymerization
	PTMG/g	MDI/g	BDO/g	PDMS/g	DCBP/g	time/min
TPU/PDMS	31.43	15.73	2.83	20	0	90
TPU/PDMS-crosslinked	31.43	15.73	2.83	20	1	40
TPU/SiR-10min	31.43	15.73	2.83	20	1	10
TPU/SiR-15min	31.43	15.73	2.83	20	1	15
TPU/SiR-20min	31.43	15.73	2.83	20	1	20
TPU/SiR-25min	31.43	15.73	2.83	20	1	25
TPU/SiR-30min	31.43	15.73	2.83	20	1	30

Table S2. The formulation for preparation of TPU/SiR TPEs with different SiR content

Sample	Formula					pre-polymerization
	PTMG/g	MDI/g	BDO/g	PDMS/g	DCBP/g	time/min
TPU/SiR-22 wt.%	31.43	15.73	2.83	15	0.75	20
TPU/SiR-28 wt.%	31.43	15.73	2.83	20	1.00	20
TPU/SiR-33 wt.%	31.43	15.73	2.83	25	1.25	20

Additional Discussion

Fig S1. The crosslinking density of PDMS-crosslinked with different crosslinking time

Table S3. Statistics of the Cross-linking density of the samples

Sample	TPU	Crosslinked PDMS	TPU/PDMS	TPU/PDMS-crosslinked
Density/g*cm^{-1}	1.07±0.08	0.83±0.11	1.02±0.15	0.98±0.06
Crosslinking density/10^{-4}*mol*cm^{-1}	3.01±0.41	5.30±0.12	3.10±0.16	7.18±0.20

Table S4. The performance data of the TPU/SiR TPEs with different pre-polymerization time

Sample	Tensile Strength (MPa)	Elongation of break (%)	Permanent deformation (%)	Hysteresis deformation (%)
10min	8.5±1.3	388±48	17.5±2.1	51.4±2.0
15min	10.2±1.2	421±43	17.8±1.8	52.2±2.7
20min	12.9±0.9	450±37	16.1±1.7	48.6±1.4
25min	10.0±1.7	436±39	17.6±2.9	53.5±1.9
30min	11.4±1.5	434±41	16.3±1.8	52.8±2.2

Table S5. The performance data of the TPU/SiR TPEs system with different SiR dispersed phase content

Sample	Tensile	Elongation	Permanent	Hysteresis

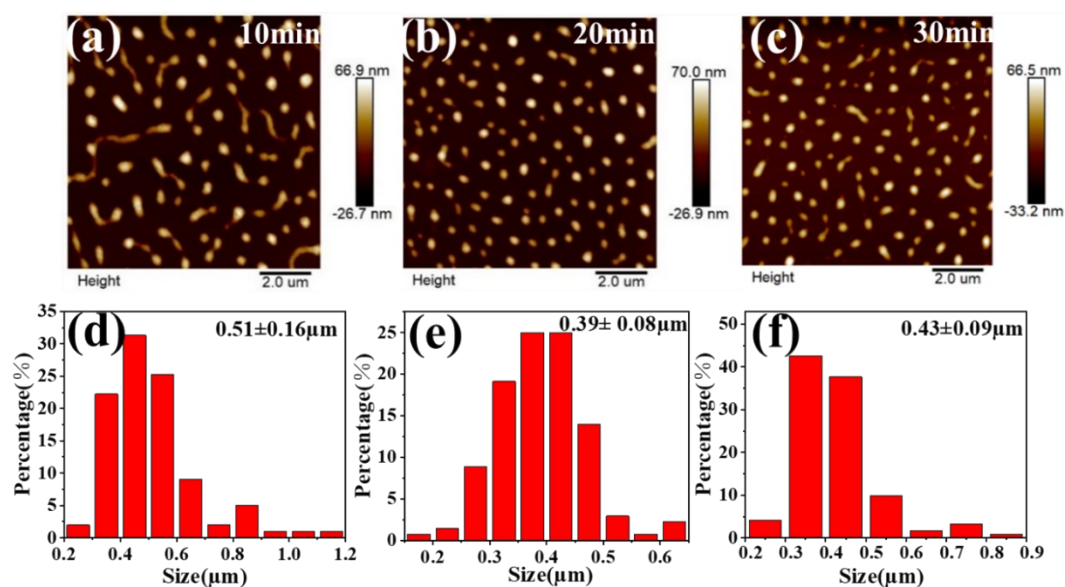|	strength (MPa)	of break (%)	deformation(%）	deformation (%)
TPU	22.9±2.2	700±49	27.6±0.3	58.1±2.7
TPU/SiR-22wt.%	13.5±0.8	528±37	15.9±1.5	51.9±0.9
TPU/SiR-28wt.%	12.9±0.9	450±37	16.1±1.7	48.6±1.4
TPU/SiR-33wt.%	10.9±1.5	434±44	19.2±2.4	55.1±1.2

Fig S2. The AFM diagram TPU/SiR TPEs with different pre-polymerization time (a) 10 min (b) 20 min (c) 30 min; and the diagram of particle size distribution (d) 10 min (e) 20 min (f) 30 min

Fig S3. The AFM height images (a, b, c), modulus images (a', b', c'), and TPU hard segment size distribution of the samples (a", b", c"): (a, a', a") pure TPU; (b, b', b") TPU/PDMS; and (c, c', c") TPU/SiR-28 wt.%
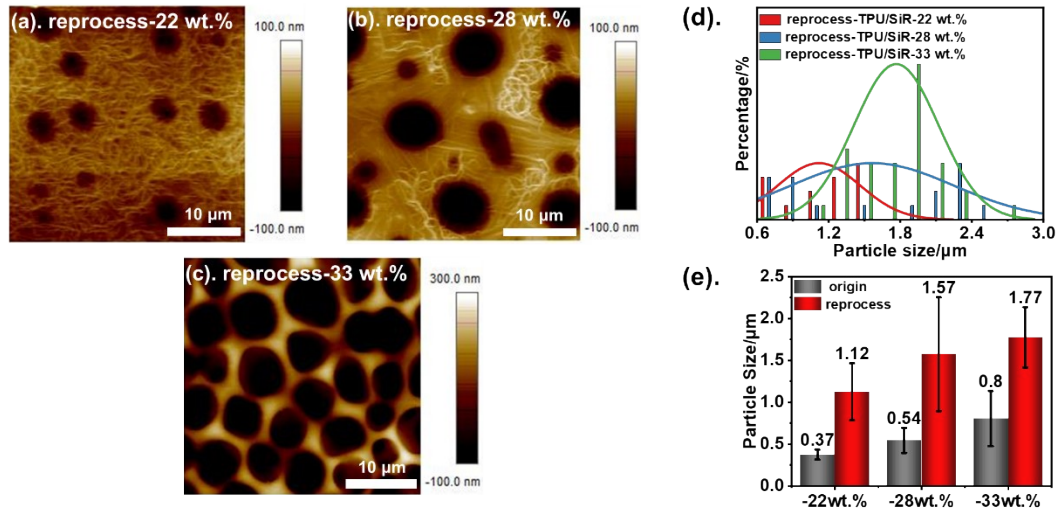

Fig S4. The AFM image of TPU/SiR TPEs with different SiR dispersed phase content after repetitive processing: (a) TPU/SiR-22 wt.%; (b) TPU/SiR-28wt.%; (c) TPU/SiR-33 wt.%; (d) the size distribution diagram and (e). the comparison of SiR particle size before and after repetitive processing of TPU/SiR TPEs with different SiR dispersed phase content.
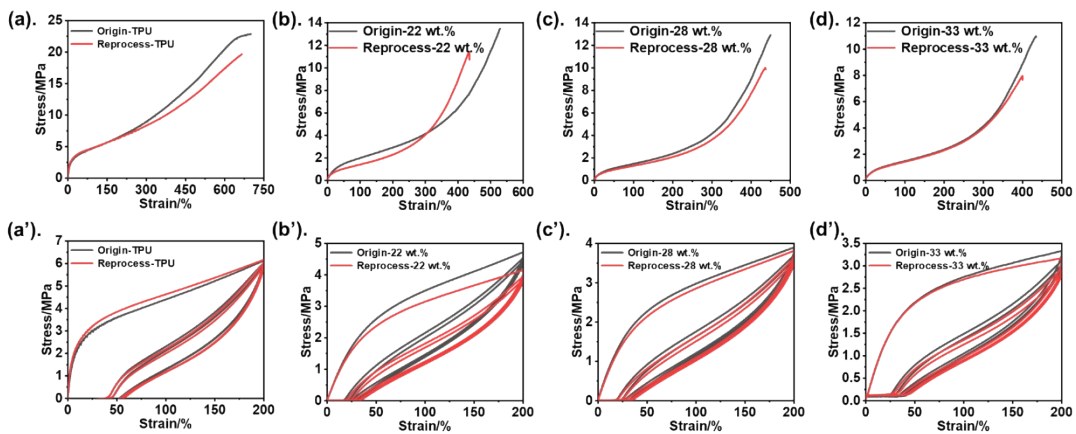

Fig S5. The Pure and TPU TPU/SiR TPEs samples with different SiR dispersed phase: (a, b, c, d). mechanical properties; (a', b', c', d'). tensile recovery properties.
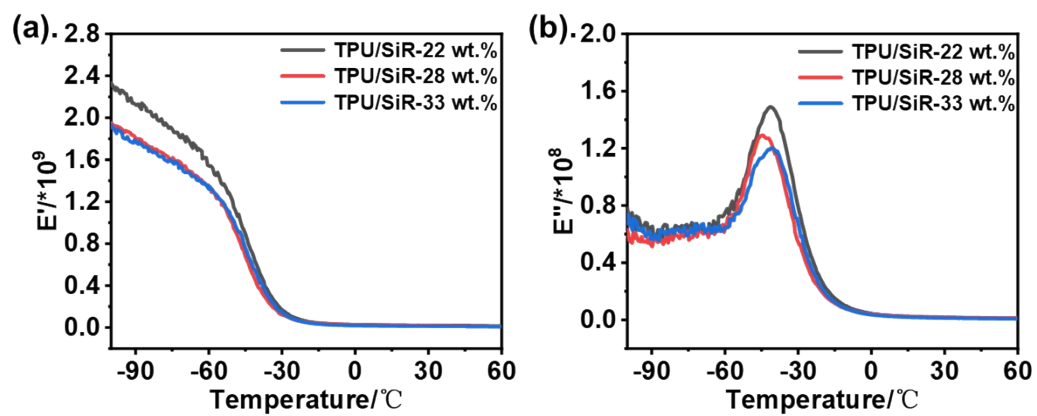
Fig S6. The TPU/SiR TPEs sample with different SiR dispersed phase content properties of (a) the storage modulus-temperature curves; (b) the loss modulus-temperature curves.